

# ICXOM23

23rd International Congress on  
X-ray Optics and Microanalysis

September 14-18, 2015

Brookhaven National  
Laboratory

Long Island, New York



Oral  
Presentation  
Abstracts



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International Conference on X-ray Optics and Microanalysis - ICXOM 23

Conference Agenda

Session title
Keynote lecture
Invited talk
Contributed talk
Breaks
General event

## X-ray Fluorescence Microscopy: Advances and Unique Opportunities

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Scanning probe microscopy has had tremendous impact on the scientific community over the past decade, addressing extremely broad and highly relevant scientific questions. To a large extent, this impact was enabled by the development of better nanofocusing, better nanopositioning, improved detector technology and better integration. X-ray fluorescence microscopy (XFM) has been one of the main beneficiaries of these developments, with the ability to image and probe the chemical state of metals present only in trace quantities. For example, in the life sciences, metals play a fundamental role in all known life forms and are increasingly recognized as having a critical impact on human health both in their natural occurrence, via therapeutic drugs, and in diseases such as Alzheimer's or Wilson's disease. In the energy sciences, for example, metal impurities and contaminants in photovoltaic materials can limit device performance, in chemistry the active components in catalysts are typically metals.

Hard x-ray fluorescence microscopy is an ideal technique to map and quantify trace element distributions in these systems. It provides attogram sensitivity for transition metals like Cu, and Zn, combined with the capability to penetrate thick samples. At the same time, the improvements mentioned above are fundamentally changing the way experiments can be carried out, giving us the ability to more completely interrogate samples, at higher spatial resolution, with larger field of view, higher throughput and better sensitivity. In combination with lensless imaging, structural information about the specimen can now be obtained simultaneously at spatial resolutions not limited by the x-ray optics used. We will report on recent advances in x-ray fluorescence microscopy, and discuss methods we have implemented, including fast fly-scanning, methods in data analysis, correlative imaging, and X-ray fluorescence micro-tomography. We will demonstrate their application in several ongoing studies, ranging from the visualization of trace elemental content in a Zebrafish embryo (see, eg, Fig1), to the investigation of plankton to further our understanding climate change.

We will also discuss challenges and opportunities for future scientific applications and instrumentation, in particular with regards to emerging possibilities with diffraction limited storage rings.

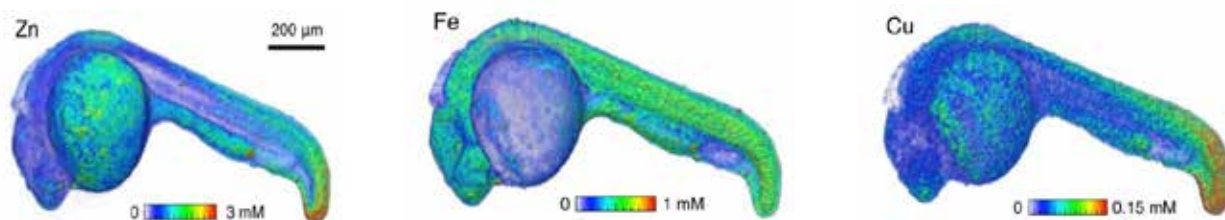


Fig.1: 3D rendering of elemental content (Zinc, Iron, Copper) of a Zebrafish embryo, reprinted from [1].

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## Chemical Speciation Imaging at Environmentally Relevant Concentrations using X-ray Fluorescence Microscopy

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### Abstract

X-ray fluorescence microscopy (XFM) [1] can be used for elemental and chemical microanalysis across length scales ranging from millimeter to nanometer. XFM is ideally suited to quantitatively map trace elements within whole plant and other biological specimens, environmental and soil samples. The elemental sensitivity of the X-ray fluorescence probe provides valuable information in a diversity of environmental sciences, and the high penetration of hard X-rays enables measurement of whole cells, tissue sections and a diverse range of environmental samples with a minimum of preparation.

Rapid advances in X-ray fluorescence detection methods such as the Maia detector [2, 3, 4] now enable high definition images approaching megapixel per minute rates. The ability to rapidly acquire 2D images enables 3D information such as fluorescence tomography to be obtained in realistic times. Chemical speciation (valence) imaging (CSI) is a technique where the third dimension is spectroscopic detail [5]. CSI results in an X-ray Absorption Near Edge Structure (XANES) spectra from the X-ray fluorescence signal at each pixel in the spatial image.

Fitting of spectra per pixel with incident X-ray energy tracking and end-member phase decomposition has recently been developed in GeoPIXE software using the Dynamic Analysis method [6, 7, 8].

CSI has been demonstrated at the Australian Synchrotron XFM beamline [1] with micron resolution and moderate definition (10K pixels) across a diverse range of sciences and applications from environmental chemistry [9] to arsenic toxicity in crop production [10]. Studies probing and optimising the efficiency and sensitivity of CSI to achieve measurements at environmentally relevant concentrations will be presented.

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## X-ray Fluorescence Microanalysis of Speleothems using Synchrotron Radiation

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X-ray fluorescence analysis (XRF) is well suited technique for investigations of the properties of geological objects. The speleothems (stalactites, stalagmites and pearls), exhibiting annual variations of trace elements concentrations, can be effectively used for monitoring of the climate changes in the past, if the XRF technique is combined with micrometer-sized intense photon beam excitation. In fact, it was demonstrated that x-ray fluorescence excited by focused microbeam of synchrotron radiation can be successfully used to study the climate changes [1].

Here we report on the x-ray fluorescence studies of stalactites and pearls samples taken from the karst “Paradise” cave, Central Poland, excited by focused x-ray photons of synchrotron radiation at the ID21 beamline of the ESRF, Grenoble. The excited x-rays were measured both by an energy-dispersive silicon drift detector (SDD) and a flat crystal wavelength-dispersive spectrometer (WDS) equipped with a polycapillary x-ray optics [2]. The WDS, operating with ADP(101), Si(111) and Ge(220) crystals, covers the x-ray energy range 1.5-6.5 keV with energy resolution 3-30 eV. The properties of this spectrometer were studied in details using ray-tracing simulation technique. The benefits of better energy resolution of the WDS, as compared to the SDD detector, for investigations of the geological samples are discussed in context of saturation and Raman scattering effects. The 1D and 2D XRF scans were measured for stalactite and pearl samples with a micrometer lateral resolution yielding the maps of concentration of six light elements between Si and K, which exhibits the annual variations. Finally, the results are discussed in context of monitoring the long-term climate changes in the past.

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## High-resolution subcellular imaging at the ESRF new nanoimaging beamline: deciphering intracellular targets of anticancer drugs in breast cancer cells

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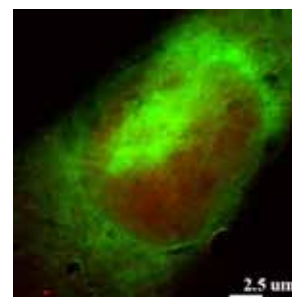
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### Introduction and Objectives

The new state-of-the-art beamline ID16A-NI at ESRF offers unique capabilities for X-ray imaging at nanometer scale delivering a highly coherent, very intense nanofocused beam ( $> 5 \cdot 10^{11}$  ph/s at  $\Delta\lambda/\lambda \sim 10^{-2}$ ) at high energies ( $\sim 20$  nm at 17 keV). It is particularly well suited for the investigation of biological samples at high resolution, e.g. the detection and quantification of trace elements<sup>[2]</sup>, such as metals in metal-based drugs in cancer treatment. Triple negative breast cancer tumors, responsible for a high rate of mortality, are a major challenge for breast oncologists since no targeting therapy is currently available for them. Jaouen's group has developed<sup>[1]</sup> ferrocenyl metal-based drug candidates that can target both hormone-dependent and independent breast cancer cells at low nM range showing encouraging anticancer effects. Our aim is to identify the targeted intracellular compartments where these compounds are active as a main step towards explaining their action mechanisms.

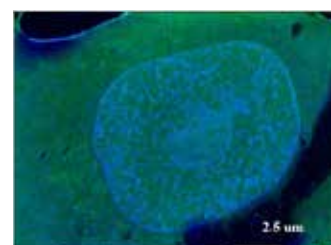
### Results and Discussion

Osmium derivatives of the ferrocenyl based drug were imaged in MDA-MB-231 breast cancer cell line using both X-ray fluorescence and phase imaging. Various sample preparation techniques were explored. First chemically fixed cells were imaged in 2D (fluorescence at 50 nm and phase imaging at 10 nm pixel size) to reveal the quantitative elemental distribution. Then we imaged 200 nm thin sections prepared by high pressure freezing for fixation followed by cryo-substitution and resin embedding. Finally X-ray fluorescence tomography scans of the entire cells were performed at 150 nm step size.



### Conclusions

The 2D fluorescence maps of entire cells consistently revealed a pattern of high Os concentration alongside the nuclear membrane, localization further confirmed by the 3D fluo-tomography data. This work confirms the potential to reveal structural information at unprecedented resolution.



Fluorescence maps of breast cancer cells at 50 nm pixel size. Up: Zn (red) and Os (green). Down: 200 nm section, Cl (green), P (blue)

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## Synchrotron radiation-induced microXRF and XANES spectroscopy to study pollutant materials in soils and assess phytoremediation strategies

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A full understanding of toxic element behaviour in the environment ultimately depend on molecular-scale structure and properties [1]. In this sense, spatially resolved Synchrotron Radiation microXRF (SR-μXRF) and XAFS allow for the study of molecular-level processes occurring at critical boundaries in environmental sciences.

In this work, the mineralogical composition and spatial distribution of Pb and Sb species in corroded ammunitions are reported to understand how they react in the soil, influencing the bioavailability and contamination risk. Furthermore, the phytoextraction technology uses plants to extract toxics metals from contaminated soils and accumulate them in the harvestable parts of the plants, which can then be removed from site. In this sense, SR-μXRF analysis is essential to examine the spatial distribution of Pb and other elements in different parts of plants in order to know its capability for incorporating these elements.

SR-μXRF elemental maps of Pb and Sb were performed at the D09B XRF Fluorescence beamline of the LNLS on selected samples focused on process like (litharge → hydrocerussite → cerussite) going to more stable phases that immobilizes Pb in soil [2]. XANES measurements at the Sb L-edges were carried out at the D04A SXS Soft X-ray Spectroscopy beamline of the LNLS in order to identify its oxidation states in crust. For plants, the experiments were conducted in *Lolium perenne* sp., grown in soil contaminated with Pb, and in hydroponics crops exposed to lead at industrial and basal levels. SR-μXRF measurements were performed *in situ* on different parts of the plant (roots and leaves) and in living conditions.

A positive correlation between Sb and Fe was detected in crust material measured in the outer rim of the weathered bullets due to Sb adsorption to Fe oxyhydroxides of soil. It was also observed a spatial correlation between Sb and Cu and between Sb and Zn in crust. Results from XANES showed that the main species found in all samples was Sb<sup>5+</sup> (Sb<sub>2</sub>O<sub>5</sub>) followed by metallic Sb. For plants, the results showed the hydroponics crops of *L. perenne* sp. can extract and translocate Pb from the ground to the leaves more effectively than plants grown in contaminated soil, where lead mainly stayed in the root. In addition, a spatial correlation between Pb, S and P distributions was observed.

Some conclusions about the current results as well as future activities will be sketched.

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## Recent Developments and Achievements at the TwinMic Spectromicroscopy Beamline of Elettra Synchrotron

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Characterisation and exploiting properties of complex materials with high spatial resolution requires the deployment of multidisciplinary techniques and expertise. Soft X-ray microscopy, combining imaging and spectroscopy at sub-micron scales, has already been recognised as a powerful technique proving both morphological and chemical information. The TwinMic microscopy station [1] operated in the 400-2200 eV energy range at the Elettra synchrotron has been attracting different scientific community, from Life Sciences to Cultural Heritage and Material Science, thanks to its complementary imaging capabilities (brightfield and phase contrast) with spatial resolution down to sub 20nm with special CDI methods, combined with low energy X-ray Fluorescence (XRF) [2, 3] and X-ray absorption microspectroscopy. Unique feature is that the developed low energy XRF system enables monitoring light elements down to B.

The presentation will use selected representative results to illustrate the recent achievements in the fields of neuroscience [4], nanotoxicology [5], clinical medicine [6,7], environmental science [8] and electrochemistry [9]. The progress in implementation of novel TwinMic imaging modalities for pushing the lateral resolution has recently been demonstrated by ptychography experiments with biological samples [10, 11]. Finally the first results of an on-going low energy XRF system development will be presented and discussed [12].

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# Table-top soft x-ray microscopy with a laser-induced plasma source

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## Introduction and Objectives

The spectral range of the ‘water window’ ( $\lambda = 2.3$  to  $4.4$  nm) represents a highly interesting regime for studying carbon-based specimen, due to a 10 times higher absorption of carbon compared to oxygen and water. This opens up a variety of applications, e.g. high resolution microscopy and near-edge x-ray absorption fine structure (NEXAFS) spectroscopy. These studies are typically conducted at synchrotrons; however, as the interest in imaging techniques and surface sensitive chemical analytics is growing, there is also a considerable demand for compact lab-based soft x-ray sources.

## Results and Discussion

Making use of the long-term stable and nearly debris-free laser-induced plasma from a pulsed nitrogen gas jet target, an extremely compact soft x-ray microscope operating in the ‘water window’ region at the wavelength  $\lambda = 2.88$  nm was installed [1]. With this microscope structures with a size of about 50 nm can be resolved (see Fig. 1).

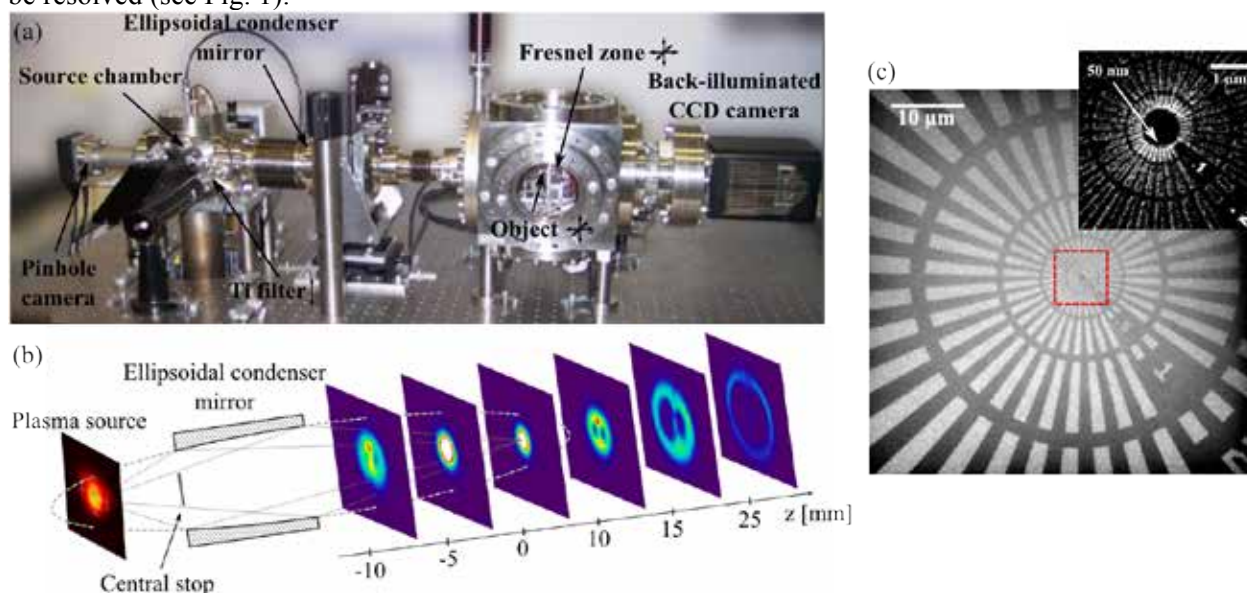


Figure 1: (a) Photograph of the table-top soft x-ray microscope. (b) Spatial intensity profiles of soft x-ray radiation at 2.88 nm for different positions along the optical axis behind the ellipsoidal condenser mirror. (c) Soft x-ray micrograph of Siemens star recorded at 2.88 nm (magnification 250x, effective pixel size 52 nm, 18 000 pulses, and exposure 60 min). The inset shows the central part of the Siemens star recorded separately at magnification 500x.

## Conclusions

In this paper, an overview on the latest results using the table-top soft x-ray microscope is given including a brief description of the imaging performance of the microscope as well as a compilation of micrographs from different sample systems.

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## Microdiffraction mapping using versatile AFM and high resolution X-ray scattering setup

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### Introduction and Objectives

As the dimensions in nanoelectronics approach an interatomic scale, the assessment of individual nanostructures becomes crucial in fabrication process of reliable devices. The shape and strain distributions in the nanostructure and at its interfaces are the parameters which influence the performance of a device [1]. X-ray scattering methods are well established for *in situ* and *ex situ* studies of shape and strain in nanostructures [2]. Using highly brilliant synchrotron sources, the coherent X-ray scattering techniques can be applied for non-destructive access to the interior of a nanostructure [3]. At the same time, the Atomic Force Microscopy (AFM) is one of the microscopy techniques to study surface morphology with nanometer resolution. Furthermore, by manipulating the AFM tip it is possible to apply local external stress to individual nanostructures and thus study their elastic properties [4]. Combination of X-ray scattering methods and AFM in one instrument enables to study locally strain fields in epitaxial nanostructures upon applying an external mechanical load by the AFM tip. This opens new research perspectives in the field of nanoscience, especially when the elastic properties of nanoscale objects are addressed.

### Results and Discussion

Building on existing experience [4] we have implemented a standard AFM system with an optical cantilever feedback which ensures stable and controlled force on the sample surface, thus avoiding the tip oscillations inherent to a tuning fork AFM setups. The AFM setup is designed to be used in combination with synchrotron X-ray beam in scattering and diffraction geometries [5]. Proof-of-principle measurements were performed at the P10 coherence beamline of the PETRA III synchrotron at DESY, Hamburg [6]. Calibration measurements in grazing incidence small angle scattering (GISAXS) geometry were carried out using Si grating sample with 300 nm pitch. Individual spintronic Fe/MgO/GaAs microring structures were studied using the described AFM sample environment. 2D diffraction maps were collected using microfocused X-ray beam and 2D pixel detector Pilatus 300K. Shape details were evaluated from GISAXS data. Elastic strains in individual structure were studied in grazing incidence diffraction (GID) geometry as induced by external mechanical stress applied by the AFM tip.

### Conclusions

Design, operation and example applications using the versatile AFM sample environment implemented at the synchrotron beamline will be presented.

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## X-ray Spectroscopy with Very High Spatial Resolution at NSLS-II

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The National Synchrotron Light Source II is a synchrotron radiation source of extremely low emittance, ideally suited for experiments in need of coherent radiation. It provides an ideal platform for sub-micrometer focused beam instruments. The Sub-micrometer Resolution X-ray spectroscopy beamline (SRX) has been developed specifically as an X-ray fluorescence analytical probe utilizing Kirkpatrick-Baez (KB) focusing mirrors. The scientific emphasis is the study of complex systems with chemical heterogeneity at sub-micrometer and sub-100 nm length scales. The beamline design provides X-ray spectroscopy capabilities in the energy range from 4.65keV to 23keV. The SRX beamline is one branch of a canted sector at NSLS-II, with a second undulator planned as a future upgrade to serve as an independent light source for another beamline, the X-ray fluorescence nanoprobe (XFN). The XFN design envisions a complementary beamline to SRX that will access lower X-ray energies (2-15 keV) and will utilize Fresnel zone plate optics. SRX uses two sets of KB mirror optics for focusing, a high photon flux set that will deliver more than  $10^{13}$  phot/sec in a sub-micron sized spot and a high spatial resolution set that will deliver a focal spot size of less than 100nm but at a lower flux of approximately  $10^{11}$  phot/sec. A highly customized horizontally deflecting double crystal monochromator was chosen to provide maximum beam stability while simultaneously providing very high spectral and spatial resolution. The energy range covered by the SRX beamline will allow for X-ray absorption spectroscopy experiments starting at the K-absorption edge of titanium and extending through the K-edge of rhodium. The photon flux SRX delivers in a submicron-spot, ultimately combined with the use of new energy dispersive detectors like the MAIA, will open new possibilities for spectroscopic analysis of major and trace elements in natural and synthetic materials, X-ray fluorescence imaging of their distribution both in two and three dimensions utilizing tomographic methods, and, concurrent micro-diffraction measurements. Diffraction imaging experiments will be developed as well. A detailed description of the SRX beamline, including results from commissioning and first experiments will be presented.

**Transforming data into information: Scientific computing for synchrotron research**

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*Submitted to: 23rd International Congress on X-ray Optics and Microanalysis (ICXOM23)*

Data volume and complexity at synchrotron facilities are increasing at a fast pace. The translation of data to information is getting more difficult due to the inability of researchers to fully comprehend the underlying analysis approaches and systems required for data processing. Even today it is virtually impossible to analyze raw data through conventional approaches without any significant investment in computing hardware or software. Moreover, future experiments will typically involve multiple detectors to measure different phenomena simultaneously providing heterogeneous data streams. All of these factors necessitate the need for advancements in scientific computing practices to match new experiments and detector technologies. In this talk, I will briefly summarize the current data-intensive science problems at synchrotron facilities; review the technical developments; outline the current status as well as future needs and directions in scientific computing for synchrotron-based research.

## Quantitative High Definition XFM Element Imaging using Maia: the Challenge of Major Composition Contrasts

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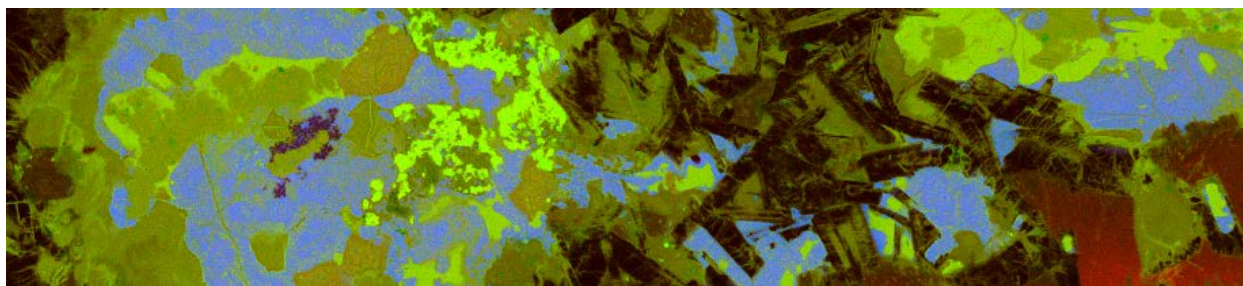
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### Abstract

Extreme contrasts in sample composition cause dramatic variation in X-ray fluorescence microscopy (XFM) yields due to self-absorption. Without *a priori* knowledge of spatial composition details, the projection of element images from XFM data using the Dynamic Analysis (DA) method [1,2] in the GeoPIXE software assumes a uniform composition and background shape. Our present approach is to then apply an iterative matrix (composition) correction. But this does not account for changing background shape and scattering and X-ray relative intensities evolving spatially with composition.

The XFM beamline [3] of the Australian Synchrotron equipped with a Maia detector [4,5], uses DA to process event data in XFM imaging and tomography at up to  $\sim 3 \times 10^7$  events/s in the Maia FPGA processor for real-time imaging or  $\sim 10^8$  on a performance desktop, corresponding to  $\sim 10^4$ - $10^5$  pixels/s and images up to  $\sim 100$ M pixels. The challenge is to improve the method while maintaining processing speed.

A new method, applied in a second pass, uses an end-member phase decomposition obtained from the first pass, and DA matrices determined for each end-member, to re-process the event data with each pixel treated as an admixture of end-member terms. This approach better tracks spatially complex samples as encountered in geological and environmental research (e.g. Fig. 1) while still benefitting from the speed of DA. This paper describes the method and illustrates how the enhanced accuracy of spectral deconvolution improves imaging of challenging materials. Direct spot comparisons with electron probe microanalysis are used to illustrate the accuracy of the method.



**Figure 1:** Maia RGB image (Cr Fe Cu, 7401 x 1650 pixels; portion of 7401 x 4601 pixels, 14.8 x 9.2 mm) of a complex geological sample acquired at the XFM beamline, illustrating challenging compositional variation.

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## PyXRF: X-ray Fluorescence Analysis Package

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A sophisticated fluorescence analysis package (PyXRF) has been developed for NSLS-II X-ray Fluorescence Microscopy beamlines, such as HXN and SRX. This package contains a high-level fitting engine, comprehensive command-line/GUI design, rigorous physics calculation and a powerful visualization interface. PyXRF offers a method of automatic elements finding, so users do not need to spend extra time selecting elements manually. Moreover, by using PyXRF we can observe the result of forward calculation at real time while adjusting input parameters. This will help us perform sensitivity analysis, and find an appropriate initial guess for fitting. Furthermore, we also create an advanced mode for professional users to construct their own fitting strategies with a full control of each fitting parameter. A convenient I/O interface was designed to obtain data directly from NSLS-II experimental database. We also discuss some fitting results from latest experiments at NSLS-II.

## Automatic Processing of Multi-modal Chemical Tomography Datasets

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Biological and material science problems are often investigated by a single microscopy/ spectroscopy technique. However, such investigations using a single technique often yield deeper questions that require further investigation. The continuing development of chemical tomography techniques, yielding spatially resolved information on e.g. elemental and phase distribution, is able to provide a more detailed picture of the nature of a material than the corresponding bulk measurements.

One example of a system that benefits from multi technique analysis is the determination of the active state of a catalyst. The physicochemical state of a material is a key factor in determining catalytic activity and selectivity, however rarely are such materials structurally or compositionally homogeneous. The use of X-ray fluorescence computed tomography (XRF-CT) is able to identify the location of the active state, but not its crystal structure or chemical state. For this, the collection of X-ray diffraction computed tomography (XRD-CT) or X-ray absorption near edge structure computed tomography (XANES-CT) is required[1]. These datasets are collected in parallel, not only reducing the duration (and hence required dose) of the experiment, but also guaranteeing the sample was in the same state for each measurement, something not possible in a dynamic industrial catalyst. This ultimately provides data for correlative image processing.

Processing time is a major consideration when designing an experiment to collect high quality data from such a multi-modal experiment, since the number of processing hours soon exceeds that which is viable for a user to spend processing them. This also detracts from the possibility of real-time analysis as the experiment progresses, so users are often left until they leave the beamline to assess how successful the experiment was. Ideally, the user would be able to view the reconstructed data as the experiment progresses, enabling them to guide the experiment on the fly.

Big data pipelines are already having a significant impact on the data analysis world outside of science, but are limited to low dimensionality data-sets. Savu[2] is a python based data processing pipeline developed at DLS that is targeted specifically at the higher dimensionality of scientific data-sets. Scientists can write simple plugins to the pipeline that can then be applied in a modular manner to the data to reduce it to its final state dramatically improving the processing time of the data.

We present the results of benchmarking the performance improvement versus a standard, user-based processing chain for this example data-set, demonstrating a dramatic increase in processing time and provide scope for future improvements as we push computationally intensive data processing towards real-time feedback.

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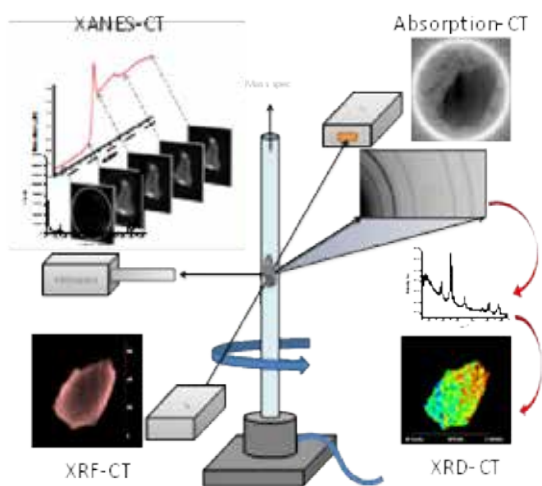


Figure 1: The experimental set-up for a multimodal absorption, XRF, XANES and XRD contrast CT, measurement of an industrial catalyst.

## nm-scale Spatial Resolution X-ray Imaging with MLL Nanofocusing Optics: Instrumentational Challenges and Opportunities

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The Hard X-ray Nanoprobe (HXN) beamline at NSLS-II has been designed and constructed to enable imaging experiments with unprecedented spatial resolution and detection sensitivity. The HXN X-ray Microscope is a key instrument for the beamline, providing a suite of experimental capabilities which includes scanning fluorescence, diffraction, differential phase contrast and ptychography utilizing Multi-layer Laue Lenses (MLL) and zoneplate (ZP) as nanofocusing optics. During this presentation, different phases of the X-ray microscope development process will be reviewed. Various prototype systems designed and constructed prior to completion of the HXN-microscope will be discussed. First data demonstrating  $\sim 15 \times 15$  nm spatial resolution imaging using MLL optics will be presented. We will discuss instrumental challenges associated with high spatial resolution imaging and will outline future development plans.

## Development of a soft X-ray ptychography beamline at SSRL and its application in the study of energy storage materials

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Ptychography is an emerging high resolution coherent imaging technique which can improve the resolution of scanning transmission X-ray microscopy (STXM) by over ten-fold. Development of this capability is underway at SSRL to establish sub-5 nm resolution in situ ptychography with near-edge X-ray absorption fine structure (NEXAFS) imaging. This is being achieved via an upgrade of the current soft X-ray STXM chamber on beamline 13-1, involving the installation of an area detector and an interferometer system for high precision sample motor control. The undulator source on beamline 13-1 provides the spatially and temporally coherent X-ray beam required for ptychographic imaging in the energy range 500 – 1200 eV. This energy range allows access to the oxygen chemistry and the valence states of 3d transition metals found in energy storage materials, making soft x-ray ptychography a particularly powerful tool to study the chemical states and structure of battery materials at relevant length scales. The implementation of in situ ptychographic imaging can therefore provide a wealth of additional information on battery operation and failure, with measurements possible at comparable imaging resolutions to TEM with a lower radiation dose. The development of this in situ ptychography capability will be described, along with its application to the study of energy storage materials.

## Strategies for Cost Minimization in X-ray Microscopy

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The clear trend in the advancement of x-ray user facilities is a steady increase in the spectral brightness of the photons produced by their sources. Practically, very bright sources benefit the end user by delivering more photons per unit phase space, which translates directly into higher flux densities in focal spots, e.g., in x-ray microscopy, and spectral windows, e.g., in inelastic spectroscopy. At the Sub-micron Resolution X-ray Spectroscopy (SRX) beamline<sup>1</sup> of NSLS-II, this capability will be exploited to provide not only world-class scanning transmission and emission spectromicroscopy, but can also be leveraged to increase spatial and temporal resolution through novel techniques that require high spectral brightness from first principles. Since these advanced techniques are not always easily implemented, the best use of the high-spectral-brightness source will depend upon the goals of the individual experiment; therefore, extracting

Using the design parameters and recent results from SRX, we will discuss the applicability and limitations—in both the spatial and temporal domains—of various advanced imaging methodologies, including coherent imaging and differential phase contrast methods. Simulations of instrument and data collection performance will be presented.

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## The Color X-ray Camera – Basics and Applications of a 2D X-Ray Detector

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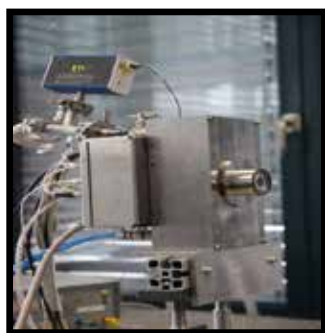
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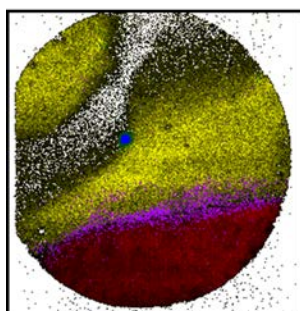
The Color X-ray Camera CXC or SLcam® is an energy-resolving X-ray camera capable of energy- and space-resolved measurements. It consists of a high-speed CCD detector coupled to a polycapillary optic that conducts the X-ray photons from the probe to distinct pixels onto the detector.

The camera is capable of fast acquisition of spatially and energy resolved fluorescence images. A dedicated software enables the acquisition and the online processing of the spectral data for all 69696 pixels, leading to a real-time visualization of the elements distribution in a sample. It was developed in a joint project with BAM, IFG Berlin and PN Sensors.

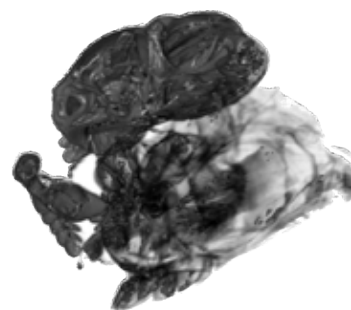
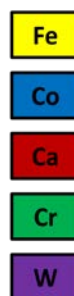
In this contribution we will mainly discuss the use of the CXC at our beamline, the BAMline at BESSY II and imaging applications of the CXC from different areas, like biology and archaeometry. Additionally new developments for the use of the detector without optics, like wavelength dispersive detection or 1shot-XANES, will be presented.



The Color X-ray Camera



Trace element distribution in bone



3-D reconstruction of a hornet

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## The EIGER Detector Systems for the Swiss Light Source

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### Introduction and Objectives

EIGER<sup>[1]</sup> is a single photon counting hybrid pixel detector developed at the Paul Scherrer Institute (PSI) for X-ray detection in the energy range from a few to 25 keV. EIGER has been specifically designed for synchrotron applications, such as coherent diffraction imaging and ptychography<sup>[2]</sup>, X-ray photon correlation spectroscopy<sup>[3]</sup>, small and wide-angle X-ray scattering and protein crystallography. EIGER features a small pixel size (75x75  $\mu\text{m}^2$ ), a high frame rate (up to 22 kHz) and a low dead time between frames (down to 4  $\mu\text{s}$ ). A 32 bit dynamic range can be obtained by data summation on-board before transferring the data to a PC.

### Results and Discussion

An EIGER module consists of a total of 500 kpixels, covering an active area of 8x4  $\text{cm}^2$ . The EIGER modules are the building blocks of large area detectors: a 1.5 detector and a 9 Mpixel one are being produced for the cSAXS beamline at the Swiss Light Source at PSI. The high frame rate capabilities are preserved for the multi-module detectors due to the fully parallel data processing.

The EIGER detector is able also to detect low energy electrons in the 10-20 keV energy range. We present an experiment in which an EIGER single chip (2x2  $\text{cm}^2$ ) is operated as a detector for photo-emission electron microscopy (PEEM) as a replacement of the traditional setup with microchannel plates, phosphor screen and CCD-camera. Due to the ideal signal to noise ratio and the small pixel size of EIGER, the detector shows excellent imaging capabilities. Measurements of the electronic and magnetic properties of Fe nanoparticles validate the detector's good performance.

### Conclusions

In the presentation, the performance of the EIGER detector, first applications at synchrotrons and the progress towards the operation of large detector systems will be presented. Plans for the development of an EIGER detector, optimized for detection of electrons for PEEM applications will also be discussed.

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## Mirror-based Optical System for the 3rd and 4th Generation Synchrotron Radiation Sources

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We have established precision figuring and figure testing methods to fabricate total reflection mirrors for synchrotron radiation hard X-rays [1]-[5]. The spot size less than 30nm has been achieved at 15keV X-ray under the nearly diffraction-limited condition [6]. For the ultimate focusing of SPring-8 beam less than 10nm, we developed a compensation optical system consisting of multilayer mirrors and a novel phase compensator which can adaptively deform with nanometer precision to correct the wavefront error to satisfy the Rayleigh's quarter wavelength criterion. In the phase compensation optics system, an at-wavelength wavefront sensing method based on a phase retrieval was originally proposed and used. Achieved focal spot size was approximately  $7 \times 8 \text{ nm}^2$  [7] - [12]. In the condensation of X-ray free electron laser, we have modified and improved our fabrication methods to meet mirrors longer than 500mm which are required especially in the SACLA focusing. 1 $\mu\text{m}$  and 50nm focusing optics have already been installed and operated for the application researches [13][14].

We have also developed an advanced KB optics system consisting of 4 mirrors for an achromatic imaging which needs to satisfy Abbe's sine condition [15][16]. Performances have been confirmed to have an imaging resolution better than 50nm and a field of view as large as 20 $\mu\text{m}$ . Coming upgrade of 3rd generation synchrotron radiation facilities must enable the multiple analysis because of an extremely high throughput of each analysis due to the unprecedented brilliance. Toward the multiple analysis, focusing optics should have flexibility to provide an appropriate sized beam for each analysis. We are developing an adaptive zoom condenser to change the spot size under the diffraction-limited condition [17].

Our current works and future plans will be shown in the development of synchrotron radiation reflective optics together with some application results.

These works were partially supported by Grants-in-Aid for the Specially Promoted Research, for the scientific research (S), for promotion of XFEL research, for CREST project, and for the Global COE Program from the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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## Pushing the Limits of Hard X-Ray Coherent Diffraction Imaging in Terms of Resolution and Sensitivity

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Scanning coherent diffraction microscopy, also known as ptychography, has great potential to close the mesoscopic gap and reach into the range of length scales between 10 nm and the atomic scale. While in conventional scanning microscopy the spatial resolution is limited by the lateral size of the nanobeam and thus by the numerical aperture of the nanofocusing optic, the spatial resolution and contrast in a ptychogram depend on the coherent fluence on the sample [1]. In addition, whether a given feature in the object can be resolved or not depends on its shape (structure factor) and can vary for different features in the object [1].

Based on these considerations, the route to highest spatial resolution and sensitivity is discussed, extrapolating from current experiments. The optimal ptychographic x-ray microscope needs a source with highest possible brilliance and x-ray optics with large numerical aperture to generate the optimal probe beam. Thus, the optimal ptychographic microscope is at the same time the best conventional scanning microscope. In addition, in order to resolve weak features within an object and exploit resonant scattering to obtain chemical sensitivity [2], the background scattering needs to be minimized, requiring special data acquisition strategies.

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## Interdigitated Silicon CRLs: A Route to Full-field Hard X-ray Microscopy

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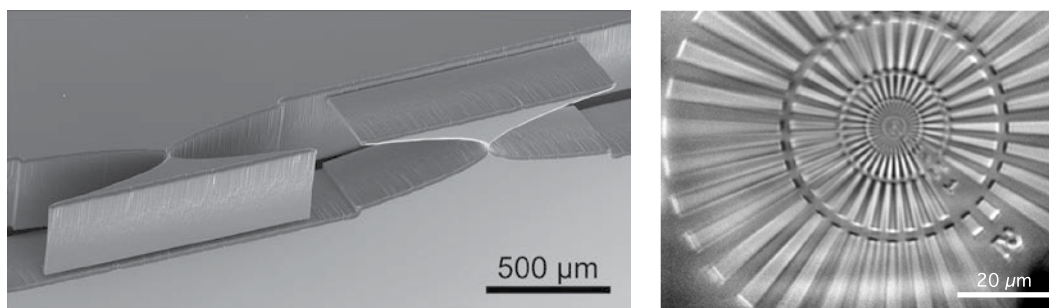
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Full-field x-ray microscopy using x-ray objectives has become mainstay of the biological and materials sciences. However, the inefficiency of existing high-resolution objectives at x-ray energies above 15 keV has limited the technique to weakly absorbing or two-dimensional (2D) samples. Here, we show that imaging objectives with significant numerical aperture and spatial resolution may be possible at hard x-ray energies by using silicon-based optics comprising 'interdigitated' refractive silicon lenslets. To achieve the 2D focusing condition, the interdigitated lenslets alternate their focus between the horizontal and vertical directions (Fig. 1a). By capitalizing on the nano-manufacturing processes available to silicon, we show that it is possible to make powerful, miniaturized imaging CRLs that minimize the inherent inefficiencies of silicon-based optics and interdigitated geometries. As a proof-of-concept of Si-based interdigitated objectives, we demonstrate a prototype interdigitated lens with a resolution of  $\approx 255$  nm at 17 keV (Fig 1b).[1]



**Fig 1.** (a) SEM image of the prototype interdigitated 2D silicon lenslet. (b) Image of resolution test chart recorded using the lens in (a) at an energy of 17 keV and magnification of 11.

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# Coherent Hard X-ray Microscopy for Mesoscopic Materials Characterization

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We present a coherent high energy X-ray microscopy branch of the multimodal instrument which is under the development at the ID06 ESRF beamline. The microscope is developed to study the wide range of mesoscopically structured materials. By employing compound refractive lenses, it is possible to combine diffraction and direct space imaging [1-4]. The diffraction pattern of the specimen formed in the back focal plane of the condenser and two-dimensional image of the object generated by objective lens in its image plane [5]. The diffraction mode is used to investigate the structure over the macroscopic distances and to orient the crystals parallel to the low index direction to perform high-resolution imaging on the local scale. The image formation relies on phase contrast due to the interference of several diffracted beams. A coherent illumination is needed in imaging mode to ensure a reasonable contrast. The coherence in terms of the angular source size determines the lens angular resolution ( $< 1 \mu\text{rad}$ ) to get high resolution diffraction patterns.

The microscope was applied for study of natural and synthetic opals, metal inverted photonic crystals and colloidal suspensions [5-6]. The combination of the direct-space imaging and high resolution diffraction provide a wealth of information on their local structure and the long range periodic order. Short acquisition times with modern area detectors allow to extend the microscope to time-resolved studies of the crystallization dynamics, response of the mesoscopic structures to external stimuli such as mechanical strain, temperature jump or temperature gradient as well as external fields.

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## Multilayer Laue lenses X-ray nanofocusing optics fabrication

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The high spatial resolution of 10nm and below needed at the next generation of synchrotron beamlines has pushed the development of nanofocusing optics. In the same time, progress in thin film deposition and nanofabrication techniques have broadened the possibilities for fabrication of new devices with larger design flexibility and control. At the crossroads of both fields, multilayer Laue lenses (MLLs) have been receiving increased attention in the last few years [1]. A MLL consists of a depth-graded multilayer which is subsequently sectioned into a high-aspect ratio structure to become a usable optic. Sectioning of multilayered structures into high aspect ratios presents enormous challenges in order to produce high quality MLLs, especially as the aperture size continues to increase. In the past few years, focusing resolution using flat MLLs has been demonstrated to approach the diffraction limit [2]. The growing demand for optics that combine high resolution, large working distance, and high efficiency for hard X-ray microscopy is now pushing the development of large aperture and wedged MLLs. In this presentation, we report our progress on the fabrication of large aperture flat and wedged multilayer Laue lenses [3].

Work supported by U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-SC0012704 for BNL and DE-AC02-06CH11357 for ANL.

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## Grating interferometry at ESRF: recent improvements and developments

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### Introduction and Objectives

X-ray grating interferometry is a multimodal x-ray imaging technique that was developed in the early 2000's [1]. During the last decade the technique has been extensively exploited for soft material characterization and biomedical applications [2,3]. Several characteristics make this technique of great interest for a wide range of applications. It simultaneously acquires three different image signals: absorption, differential phase and dark-field [4]. It enables the 3 dimensional quantification of the index of refraction of materials. Furthermore, this technique can be easily implemented at laboratory x-ray sources [5]. For all these reasons grating interferometry is of particular interest for many different fields in need of high sensitivity in the micrometer range. However, the image acquisition procedure remains quite long (~ few hours) in comparison to other x-ray imaging techniques such as propagation-based imaging (~ few minutes). This work presents an important step on the way of making the image acquisition in grating interferometry shorter and competitive with other phase contrast imaging techniques.

### Results and Discussion

In the following we will show the latest advances of the instrument at the ID19 beamline of the ESRF, which was previously only used with monochromatic x-rays with an energy bandwidth of  $\Delta E/E \approx 10^{-4}$  reached by Bragg-reflection/double-crystal monochromator Si(111). The instrument has now been characterized using x-rays with a broader energy bandwidth of  $\Delta E/E \approx 10^{-2}$ . A specific high sensitivity phase-phantom was designed for this purpose. We show the results of the measurement of this phantom and we discuss the challenges of making the imaging procedure shorter while keeping the quantitiveness and the high sensitivity of it.

### Conclusions

Grating interferometry is a unique technique to quantitatively characterize the index of refraction of different materials with a sensitivity that is not comparable to any other non-destructive technique. The use of broader energy bandwidth x-rays and higher flux can be useful to perform shorter scans. Big efforts are being made towards faster acquisition schemes that will in the near future shorten the experimental time needed.

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## Zone Plates for Nanoscale X-ray Imaging

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Fresnel zone plates are the key optical elements for nanoscale imaging of soft and hard X-rays with high spatial resolution. To date, highest spatial resolutions of about 10 nm half-pitch were reported [1-3] in zone plate based x-ray microscopy. However, conventional zone plates manufactured by planar nanostructuring processes are limited by the achievable aspect ratios of their zones. Therefore, the combination of high spatial resolution and high diffraction efficiency is still a fundamental problem in X-ray optics. Furthermore, electrodynamical simulations show that efficient, ultra-high resolution zone plate optics require 3-D shaped tilted zones to benefit from volume diffraction effects.

Our approach to realize such advanced 3-D profiles for soft X-ray focusing is on-chip stacking of multiple zone plate layers with decreasing zone radii. Additionally, on-chip stacking permits to manufacture high efficient, high resolution diffraction optics for the tender and hard X-ray energy range by multiplying the aspect ratio of the zone structures. We demonstrate triple layer on-chip stacked zone plates with an overlay accuracy of sub-2 nm which fulfills the nanofabrication requirements [4].

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## Application and Optics Concepts for Scanning and Full-field X-ray Microscopy with Multilayer Laue Lenses

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### Introduction and Objectives

Multilayer Laue lenses (MLL) are an emerging type of X-ray lenses that are fabricated by thin film deposition of multiple layers obeying the zone plate law on a smooth substrate and by subsequent micromachining such as focused ion beam milling. This technological approach results in very small zone widths down to single nanometers and virtually unlimited aspect ratios. Thus, enhancements in terms of efficiency and resolution are expected in particular for hard X-ray applications. We report (a) on recent results of the characterization of crossed and wedged MLLs using scanning X-ray microscopy, and (b) on an alternative condenser concept using MLLs for full-field X-ray microscopy with high photon energies.

### Results and Discussion

We manufactured a multilayer stack consisting of 7000 individual layers to obtain MLLs with a focal length of 6.7 mm at 10.5 keV X-ray photon energy using WSi<sub>2</sub> and Si as alternating absorber and spacer materials. From this multilayer stack, several flat and wedged [1] MLL segments were manufactured. Both types were then used for point focusing experiments at beamline ID 13 of ESRF. With focal spot sizes of 33 nm × 28 nm and 43 nm × 44 nm reconstructed by ptychography [2], both types provided nearly diffraction limited focusing for their respective apertures of 23 × 23 μm<sup>2</sup> and 15 × 15 μm<sup>2</sup> [3].

It was shown that MLLs are potential novel optics for full-field X-ray microscopy [4]. The illumination of the object plane can be optimized, if the monocapillary is replaced by a multilayer mirror based condenser. This setup is characterized by a better matching of the numerical apertures of condenser and MLL, a decent monochromatization, and the applicability of photon energies  $E > 10$  keV.

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## High-energy X-ray Focused Microbeam by using KB Mirrors and Sagittal Bent Laue Monochromator

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### Introduction and Objectives

It is proven that the high-energy X-ray diffraction in terms of atomic pair distribution function (PDF) technique is a powerful tool for studying crystalline, disordered and nano materials [1-3]. Although there is increasingly demand for high energy X-ray microbeam in the fields of material sciences and high pressure geosciences, focusing a high energy X-ray beam down to microbeam size is still a challenge.

### Results and Discussion

The double-Laue crystal monochromator (DLCM) has advantages of high angular acceptance, high photon flux and high thermal loading stability, making it well suited for providing the intense high-energy X-ray beam. The high-energy DLCM at the X17B3 beamline, the National Synchrotron Light Source (NSLS), Brookhaven National Laboratory, has a fixed energy at 80 keV for high-energy diffraction experiment[4]. However, acquiring PDFs using a large unfocused beam for high-pressure DAC experiments results in low intensity imposed by tiny sample chamber of the DAC and long-time data collection, making PDF measurement at pressures much higher than 10 GPa not yet possible [2].

In this conference, we present our recent progress in focusing the high energy X-ray beam to obtain intense microbeam by using KB mirrors in combination with sagittal bent Laue monochromator. Some examples of the compressibility properties of nanoparticles, such as n-Au, n-Ag and n-Pt, in the diamond anvil cell under quasi-hydrostatic conditions at high pressures will be presented.

### Conclusions

We have obtained intense high-energy microbeam for decent high-pressure PDF measurements by a combination of KB mirrors with the bending DLCM focusing techniques.

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## A Large Area Rapid Imaging Analysis Tool.

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The Large Area Rapid Imaging Analysis Tool, or LARIAT, has been designed to provide full-field near edge x-ray absorption spectroscopy (NEXAFS) imaging. Using the combination of a magnetic projection lens and grid-less electrostatic lenses, LARIAT is capable of collecting all electrons emitted within the field of view, leading to high quality images with a collection rate limited in many instances by the electronics readout. Raw image resolutions on the order of 5  $\mu\text{m}$  have been achieved, and can be improved by an order of magnitude using a suite of image processing tools. LARIAT will be installed on the NIST 'SST' beamline suite at NSLS II. Here we will provide an overview of the system design, some initial results from testing at NSLS, and recent updates.

## Developments in Reflective X-ray Optics for the ESRF Upgrade Programme

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An important element of Phase I of the ESRF Upgrade Programme, has been the renewal of the X-ray instrument portfolio with 19 new or refurbished beamlines. This programme has driven the development of a considerable number of new reflective optical systems. Within this upgrade programme over 35 mirror and multilayer optics systems for the new beamlines have been designed, assembled and installed by the ESRF. These devices range from high heat-load mirror systems to multilayer-based nanofocusing mirror systems and also include multilayer-based monochromators. The use of generic design principles has enabled the delivery of tailored systems to the beamlines based upon well proven solutions. In particular for the micro- and nano-focusing mirror systems this approach has permitted an optimized integration of the opto-mechanics within the end-station design allowing the use of compact and inherently stable focusing.

Critical for the success of this strategy has been the availability of in-house expertise in a variety of domains, spanning mechanical design and modelling, cooling technologies, thin-film coating methods and advanced motion and surface metrology methods. For the latter it is particularly important for the ex-situ validation of the system that, wherever possible, characterization of the optical surface can be performed not only in the unconstrained state but also in the final opto-mechanical mount.

This talk will present a selection of the reflective optics which have been implemented at these new ESRF beamlines with particular emphasis upon i) high-heat load mirror systems and ii) dynamically and statically figured mirror systems for micro- and nano-focusing. These illustrated examples will also serve to demonstrate the current level of ESRF expertise in the various key technological fields indicated above.

Phase II of the ESRF Upgrade Programme, which began in 2015, heralds the beginning of the project which will involve the replacement of the current ESRF storage ring within the existing accelerator tunnel by a new ultra-low-emittance 6 GeV hybrid multibend achromat (MBA) lattice [1]. This will offer hard X-ray synchrotron beams of unprecedented brightness and coherence. Some of the challenges and opportunities that will be offered by the new beam characteristics for X-ray optical systems will be addressed.

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## Development of bimorph X-ray nanofocusing mirrors

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### Introduction and Objectives

In recent years, several schemes have been proposed to focus intense X-ray beams into a spot with diameter <10 nm. Various optics such as mirrors, multilayer Laue lenses, and zone plates have been proposed to create nano-focused X-ray beams. Although the principle of X-ray nano-focusing was proven several years ago, routine production still remains a technological challenge. Making such beams available to the general scientific community would greatly enhance many research areas including X-ray imaging, spectroscopy, and nano-diffraction. A joint R&D program between Thales SESO and the Optics Group at Diamond Light Source (DLS) has been established to create a nano-focusing X-ray mirror based on bimorph technology previously developed at Thales SESO [1-3].

### Results and Discussion

During the first year of collaboration, a set of concepts and technologies were investigated. Two prototypes were created based on 1<sup>st</sup> [1, 2] and 2<sup>nd</sup> [3, 4] generation bimorph mirror technology. The ultrathin 1<sup>st</sup> generation prototype (150mm long) was polished flat using traditional methods. Sixteen embedded piezo electrodes permit localised correction of polishing defects and for the mirror to be bent to an extremely wide range of curvatures (~30 m concave to ~120m convex). This enables the mirror to be bent to a short radius ellipse optimised for X-ray use at DLS's Test beamline B16 [5]. A novel 2<sup>nd</sup> generation bimorph prototype was also built. This substrate is substantially thicker and its optical surface was pre-shaped to a given ellipse using traditional polishing and ion beam figuring. A number of polishing techniques were combined at different stages of mirror fabrication. In both cases, a set of wavefront sensing methods and metrology instruments were used to assist development of these two technologies [6].

### Conclusions

Two mirror prototypes, based on 1<sup>st</sup> and 2<sup>nd</sup> generation bimorph technology, have been created. Their dynamic behaviour has been characterised using X-rays and traditional visible light instruments including interferometers and the Diamond-NOM slope profilometer. Advantages and shortcomings inherent to both technologies, as well as focusing results, will be discussed during this contribution.

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## Development of X-ray multilayer gratings with high resolution and high efficiency

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High resolution X-ray diffraction optics with high efficiency are the key components to probe the structure and dynamics of matters. Combining X-ray multilayers with high quality gratings can further improve the spectral resolution and efficiency/throughput of the diffraction optics to serve the advanced applications like high resolution x-ray spectroscopy and imaging [1]. A unified analytical theory and different types of multilayer gratings are under development in our lab, particularly the blazed multilayer grating (BMG). The unified analytical theory presents consistent results with numerical methods with much faster calculation speed, which also help reveal the influence of the grating structure on its optical properties [2]. The blazed grating substrate is produced by wet anisotropic etching of a single crystalline silicon. This method can generate sharp triangular profile with atomically smooth surface which is very important for achieving the high efficiency in theory [3]. Progress in both theory and experiments will be discussed here.

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## High Energy Micro-Diffraction

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Many natural and technologically important interfaces lie deeply buried beneath their exposed surfaces. In order to access them with a non-destructive technique, an in situ probe with the potential for molecular-scale resolution is required. High-energy x-ray micro beams are the ideal probe due to their large penetration power, thereby making a wide variety of buried interfaces accessible for structural studies. The basic principle exploits the large penetration of high-energy x-rays at low angles so that only the one interface of interest is illuminated with x-rays. This reduces the number of interfaces that contribute to the scattered signal and enables the use of two general techniques, x-ray reflectivity and grazing-incidence diffraction, on interfaces that are otherwise inaccessible. It has turned out that the very small size of the incident beam, as required for small incident angles at high energies, also serves to reduce the background, giving access to molecular or even atomic resolution in most cases.

Any information encoded within the diffraction signal can be used to reconstruct 2D or 3D representations of the state of a sample providing much more physically and chemically relevant information than conventional tomographic techniques based on electron density variations alone. Thanks to recent advances in high-energy x-ray production, focusing and detection, time-resolved diffraction tomography experiments are now possible. This has been most prominently employed for the study of the evolution of industrial catalysts undergoing a thermal treatment as a route to the formation of the active phase. The obtained results constitute a new way of monitoring intricate chemistry in space and time within materials.



## Visualizing pigment distributions and their degradation products in oil paintings using a non-destructive macroscopic XRF/XRPD scanning system

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Over the past years macroscopic X-ray fluorescence (MA-XRF) has proven to be a useful tool for imaging of large cultural heritage objects, e.g. oil paintings.<sup>[1-3]</sup> The resulting elemental maps provide complementary information to methods such as X-ray radiography (XRR) and Infra-red reflectography (IRR) that are traditionally used for the investigation of these artworks, and can be used to study the distribution of various pigments on or beneath the surface. However, a main limitation of MA-XRF is that the distribution images are element-specific rather than pigment-specific. This becomes a problem for example when the pigments lead white (cerussite,  $\text{PbCO}_3$  and/or hydrocerussite,  $2\text{PbCO}_3 \cdot \text{Pb}(\text{OH})_2$ ), red lead (minium,  $\text{Pb}_3\text{O}_4$ ), Naples Yellow (bindheimite,  $\text{PbSb}_2\text{O}_6 \cdot \text{PbO}$  or rosiaite,  $\text{PbSb}_2\text{O}_6$ ) and Lead Tin Yellow ( $\text{Pb}_2\text{SnO}_4$ ) are used together, which will all contribute to the Pb-L and Pb-M distribution maps. A second drawback of this technique is the energy-dependent sampling depth for the different elements. The elemental images corresponding to low energy X-rays such as e.g. Pb-M, K-K, Ca-K show information limited to only the top surface of the painting, while for lighter elements such as, e.g. Na, Si, P no fluorescence signals are detected.

To overcome the above-mentioned limitations, a laboratory macroscopic X-ray powder diffraction (MA-XRPD) system has been constructed employing a monochromatic (Ag-K $\alpha$ ) X-ray source of low angular divergence ( $1\mu\text{S}^{\text{HB}}$ , Incoatec), delivering about  $10^7$  ph/s in a focal spot of around 100  $\mu\text{m}$  and a single photon-counting X-ray diffraction camera (PILATUS 200K, Dectris). This system allows for the simultaneous recording of XRF and XRPD distribution images from macroscopic objects with a typical lateral resolution in the 0.1 – 1 mm range and dwell times per pixel of several seconds (1-10 s).

Both in our laboratory as well as on-site, a series of measurements on oil paintings from various artists and time periods were performed that reveal meaningful pigment-specific distribution images of different crystalline compounds such as lazurite ( $\text{Na}_4\text{Ca}_4\text{Al}_6\text{Si}_6\text{O}_{24}\text{S}_2$ ), azurite ( $2\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$ ), calcite ( $\text{CaCO}_3$ ), hydrocerussite, cerussite, cinnabar ( $\text{HgS}$ ), bindheimite, etc. yielding information on both visible and covered paint layers. Furthermore, the results highlight the different usages by the artists of cerussite and hydrocerussite, both of which are found to be present in different compositions in the pigment lead white. As a final part the presence of various compounds that are believed to have formed over time as alteration products on these artworks will be discussed.

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## Orientation changes upon attachment of spider hairs investigated *in situ* using scanning X-ray nanobeam diffraction and small-angle scattering

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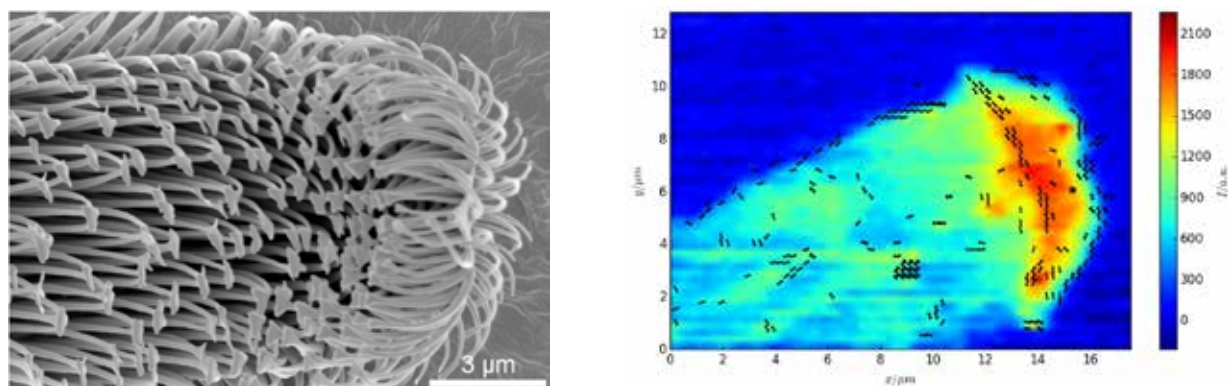
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The hairy attachment system of spiders enables these animals to walk upside-down on plants, walls, on rough and smooth surfaces, and support a multiple of the body weight without the use of glue. These outstanding biological structures comprise of pads including hundreds to thousands of specially designed hairs that are made of composite materials consisting of proteins and reinforcing chitin fibers (Fig. 1). The goal of our study was to gain an in-depth understanding of the working principle of the attachment and detachment processes of single hairs to a surface, which is likely based on the structurally determined material properties of the contact elements at the tips of the hairs. The accompanying changes of the shape of the contact elements and their self-orientation in parallel with a surface lead to the establishment of van der Waals forces between the hair tips and the substrate crucial for attachment.

Scanning X-ray nanobeam diffraction and small-angle scattering are ideal tools to reveal the structure and orientation of the smallest attachment hairs. Moreover, they can be combined with an *in situ* attachment/detachment procedure. The experiments were performed at the nanobeam of beamline ID13 (ESRF, Grenoble, France) with a beam size of 100 x 100 nm<sup>2</sup> and an energy of 14.9 keV. The tips of single adhesive hairs of the spider *Cupiennius salei* were scanned in a combined WAXS/SAXS configuration in fine detail with a resolution of 250 nm. A piezo-driven actuator was used to move the hairs relative to a thin silicon nitride window in order to attach and detach them, respectively.

Maps of the WAXS and SAXS signals allow for a detailed analysis of single contact elements (Fig. 2). Taking the dimensions of the structures with a width of about 1 µm and a thickness of less than 100 nm into account, this result is remarkable. The SAXS signals reflect the fiber orientation. Changes upon hair attachment concern those orientation aspects of the thinnest structures. Oriented WAXS signals of chitin (fiber diffraction patterns) are only found in the hair shaft 50 µm away from the tip. At the tip, amorphous rings are measured. This finding points to a complex structural embedding of the chitin fibrils in the protein matrix and to relatively small chitin aggregates. — Our results will help to improve artificial attachment devices.



**Fig. 1** (left): SEM image of the tip of a spider attachment hair; the spatulae making the contact to a surface can clearly be seen. **Fig. 2** (right): combined map of WAXS intensities (diffuse ring; color scale) and SAXS orientation (black lines) of an attached spider hair.

## Science and Development Programs at NSLS-II

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NSLS-II is the newest 3<sup>rd</sup> generation synchrotron facility in the world and started its operations in the last quarter of 2014. When fully built out, NSLS-II will accommodate more than 60 beamlines and provide high photon brightness and flux as well as excellent beam stability over a broad range of photon energies, and will enable a wide-range of cutting-edge scientific programs from materials and chemical sciences to environmental and life sciences.

In this presentation I will provide an overview of the NSLS-II scientific programs currently being developed and commissioned, and describe the suite of research and development projects in x-ray optics and instrumentation that are ongoing at NSLS-II. These projects include nanofocusing optics, nanopositioning, optical metrology, coherent imaging methods, and x-ray optical simulations. I will conclude with an outlook of the future directions that NSLS-II plans to pursue and the ongoing effort to define the next suite of beamlines to be constructed at NSLS-II.

## Correlative Multiple-length-scale X-ray Microscopy at SSRL

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The studies of complex heterogeneous systems, e.g. research in functional materials and geoscience, usually requires a suite of analytical tools that are capable of providing complementary information about material properties at different length scales. This is due to the fact that the heterogeneity of the complex system usually exists across a wide range of length scales.

X-ray microscopy offers several different imaging modalities, such as full-field X-ray microscopy, scanning X-ray microscopy, and coherent-based X-ray microscopy. These imaging modalities probe structural and chemical information at different scales ranging from macro-, micro-, meso-, to nano- scales, depending on the X-ray optics used and the configuration of the imaging systems. The pros and cons (in resolving power, sensitivity, data acquisition speed, etc.) of each individual X-ray imaging method naturally make the case that the correlative imaging is powerful and beneficial for scientific studies in many fields. In this presentation, we show our efforts to establish multiple-length-scale X-ray microscopy platform at SSRL [1-4]. We demonstrate the strength of correlative multiple-length-scale X-ray microscopy by presenting two systematic research works, one on complex heterogeneous catalysis material for petroleum refining [5] and the other one on underground formation for CO<sub>2</sub> sequestration [6]. The scientific cases serve as good examples to show the link between the macroscopic behavior and the microscopic properties at multiple-length-scales.

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## Revolutionary X-ray Microbeam Delivery System

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We are developing a revolutionary x-ray microbeam delivery system that provides about 100X higher x-ray flux density than the highest performance commercial x-ray microbeam system currently available. The new system is based on a super bright laboratory source that is designed to be substantially brighter than the brightest rotating anode x-ray source currently available, coupled with high efficacy, large solid angle, axially symmetric optics. It will also offer a substantially wider selection of characteristic x-ray energies than are available using the current x-ray source technologies. The outstanding performance of the x-ray source is achieved with patent pending x-ray source technology that incorporates the outstanding thermal and material properties of diamond as a part of the microstructured anode, creates large thermal gradients within the microstructure, and incorporates an optimized electron energy deposition profile. The source is also designed to accumulate x-rays generated from a linear array of x-ray sub-sources. In addition, it uses axially symmetric x-ray reflection optic with a large solid angle of collection from the source and customer selectable beam collimation profile. The high flux x-ray beam delivery system represents an important innovation in x-ray beam delivery technology. Its performance will enable substantial performance improvement in a wide range of x-ray analysis techniques, such as micro x-ray fluorescence (uXRF), x-ray diffraction (XRD), small angle x-ray scattering (SAXS), and total x-ray reflection fluorescence (TXRF). The design and expected performance will be presented.

## High Dimensional Diffractive Imaging with Soft X-rays at the Advanced Light Source

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The resolution limitation of scanning x-ray microscopes has recently been overcome through the development of x-ray ptychography. A ptychographic microscope operates much like a conventional scanning probe system in that the sample is scanned quickly and precisely through a focused illumination. During the scan, the sample is positioned such that neighboring positions are partially redundant and a full coherent diffraction pattern is measured at each point. Various phase retrieval algorithms have been developed which can reconstruct the complex valued transmission of the object and its illumination using knowledge of the diffraction intensities and the sample positioning [1,2]. Effective ptychographic imaging requires a high brightness x-ray beam, high speed detectors, precise and fast scanning, and high performance algorithms and computation for image reconstruction. The Advanced Light Source is leading an inter-divisional collaboration within Lawrence Berkeley National Laboratory which brings together the world's experts in each of these key areas for the development of a ptychographic imaging facility [2,3,4]. Our instrument, called the Nanosurveyor, can generate ptychographic data at 200 Hz and provide realtime imaging feedback to the user via streaming analysis algorithms run on remote, GPU based, computational resources connected by a high speed network. Using this instrument on a bending magnet x-ray source, we couple soft x-ray ptychographic imaging, x-ray absorption spectroscopy and computed tomography to map the oxidation states in a nano-particle based battery electrode at 11 nm resolution in three dimensions. Furthermore, we exploit the availability of both absorption and phase contrast in ptychographic imaging to measure the full refractive spectrum of various materials for the first time. We characterize the unique relationship between this spectrum and the achieved image resolution and show that the complex spectrum can be used to quantify chemical composition with few nanometer resolution. The Nanosurveyor instrument will be moved to a new high brightness beamline in 2016, called COSMIC, which will enable three dimensional imaging with chemical specificity, wavelength limited spatial resolution, and time resolution adequate for visualization of *in situ* electrochemical phase transformations.

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# Imaging with Coherent X-rays: From Technique Development to Single Particles

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The technique of Coherent X-ray Diffractive Imaging (CXDI) [1] utilizes algorithmic phase retrieval to generate an image of a sample using measured diffracted intensities. The achievable resolution is limited in principle only by the wavelength of the incident x-rays and the size of the detector used. It promises the ability to obtain a high-resolution image of any object, without the need for crystals. It will soon be two decades since the original demonstration of the use of coherent x-rays to image non-periodic objects [2]. Since this initial demonstration, a lot has changed in the world of x-ray science and not surprisingly, a lot of developments have taken place in the pursuit of high resolution coherent x-ray diffractive imaging.

Over the years, intense effort in algorithm development has made CXDI a fairly mature method that can now be employed in multiple fields such as biology [3] and condensed matter systems [4]. Scanning techniques have been developed allowing extended objects to be imaged. On overview of the CXDI technique and recent applications will be presented.

New x-ray sources are transforming x-ray science by enabling previously unachievable measurements to be performed. The new storage ring machines coming online will provide much increased coherent flux potentially allowing measurements that were previously signal-limited. Fourth generation light sources such as free electron lasers offer new and unique possibilities of limiting the radiation damage during the measurement [5] as well as unprecedented time resolution. The use of the CXDI technique with new x-rays sources will be presented, including time-resolved studies in materials [6] and developments towards using CXDI to image single particles, potentially single biological molecules [7].

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## Simultaneous and sequential X-ray ptychography and fluorescence microscopy at the Australian Synchrotron

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### Abstract

X-ray ptychography have seen many recent advances including sample delivery [1], data collection and analysis [2, 3], and data interpretation [4, 5]. Some of these advances have allowed water-window ptychography to be applied to hydrated specimens [1, 6] allow biological specimens to be imaged with high contrast in close to their natural environment. Other advances, such as on-the-fly scanning and rapid data processing offer improvements in the efficiency of ptychography that make simultaneous application of other scanning microscopy methods such as X-ray fluorescence microscopy experimentally viable [7]. For some specimens, the experimental requirements of these two methods overlap, and the measurements are best taken simultaneously [8]. However, for many cellular applications, the ideal conditions for each method are very different, requiring a sequential set of optimized measurements for ideal results [9]. In this presentation, we outline the case for each type of measurement and discuss recent applications of these two methods at the X-ray Fluorescence Microscopy and Soft X-ray Imaging beamlines at the Australian Synchrotron.

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## Diffraction imaging with coherent X-rays from SACLA and SPring-8

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Coherent X-rays are becoming more amenable to nano-scale imaging of weakly scattered objects. Recent operations of X-ray Free Electron Lasers (XFELs) as well as advanced synchrotron radiation sources have greatly facilitated structural investigations of specimens ranging from nanocrystals, biological cells, organelles, to macromolecular complexes. Various imaging modalities such as wet-CDI, single-shot 3D, pump-probe imaging, 3D tomography, etc, have been applied to better address the scientific issues of each systems, enriched by different characteristics from different types of light sources. We have employed coherent X-rays from SACLA and SPring-8 for imaging various specimens, and also demonstrated that the combined use of them enabled self-contained analysis of RNAi macromolecular complexes with applications not limited to. Those of our recent progresses on diffraction imaging will be introduced, and further issues related to the sample handling, signal-enhancement, etc., are discussed.

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## Three-dimensional Mapping of Strain Filed in SiGe Thin Film

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The understanding and manage of strain is of fundamental importance in the design and implementation of materials, and a key issue that determining the performance of semiconductor devices. Conventional x-ray diffraction techniques, although capable of measuring strain at very high accuracy, do not offer the spatial resolution needed for microelectronics. X-ray Bragg ptychography [1] is a new emerging technique to image strain noninvasively in an extended crystalline specimen with a resolution better than the beam size of the x-ray probe, ultimately enabling a 3D strain measurement at the nanoscale. In this work, we present a theoretical study on the effects of partial coherence, the overlap ratio and position uncertainty in Bragg ptychography, using synthetic data generated from forward simulation. The convergence of the reconstruction and the detection sensitivity of the strain field under various conditions are discussed. We also report a Bragg ptychography experiment on a SiGe thin film feature for 3D strain reconstruction by performing spiral 2D scans at different rocking angles. Reconstructed strains are compared to previously reported values and results obtained from theoretical modeling. Issues and challenges in the reconstruction are discussed as well.

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## Investigation of diffuser influence in near-field ptychography

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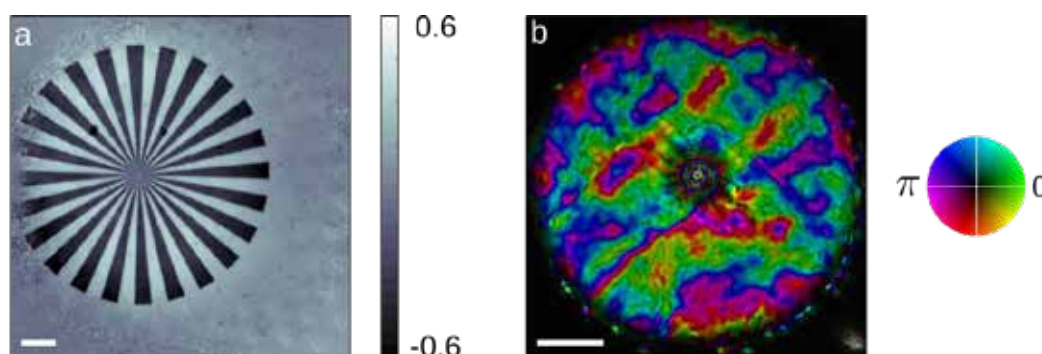
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Ptychography is a coherent diffractive imaging technique [1] utilizing a scanning transmission microscopy setup where overlap redundancy of the incident illumination between adjacent scan points is used to reconstruct iteratively both the object's transmission function and the scanning illumination profile [2]. Near-field ptychography, a novel modality of this technique using Fresnel diffraction patterns, was recently demonstrated [3]. The method has been applied to strongly phase shifting samples [4] and in conjunction with nanotomography [5]. Benefits of near-field ptychography include large fields of view and absence of wavefront corrections since wavefront imperfections are explicitly required by the reconstruction approach. In fact, additional distortions are introduced into the scanning beam with the aid of a diffuser to generate more diversity. The influence of the diffuser on the reconstruction process has yet to be investigated and is at the center of our research as we further explore the possibilities of this new technique. We present the results of a diffuser study carried out at the coherence beamline I13-1 at Diamond Light Source. A Siemens star test pattern was imaged with different diffuser materials in the beam and their influence on the reconstruction process was investigated. In addition, the effect of translating the diffusers along the optical axis was examined. Our findings are supplemented with visible light measurements. We acknowledge financial support through the European Research Council (ERC, starting grant "OptImaX").



**Figure:** Reconstructed phase shift of a Siemens star test pattern (a) and corresponding complex-valued illumination function distorted by a weak cardboard diffuser (b). Scale bars indicate 20  $\mu\text{m}$ .

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## Recent Developments in Environmental High-Resolution Transmission Electron Microscopy

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Transmission electron microscopy has proven to be a powerful tool to measure composition, chemistry and internal structure at the nanoscale and below. The two transformative developments in electron microscopy in the last two decades are (1) the emergence of aberration-corrected lenses that allow for unprecedented spatial and spectral resolution and (2) the rapid advances in *in situ* capabilities for observations of dynamic phenomena.

Recent and rapid developments of *in situ* transmission electron microscopy (TEM) has demonstrated it to be a transformative tool to gain unique dynamic processing/structure/property relationships of nanomaterials. Of particular interest are the structural changes occurring under “real” environmental conditions observable by environmental TEM (ETEM). The ETEM allows for dynamic studies for fundamental, atomic-level understanding of surface chemical reactions, such as oxidation and heterogeneous catalysis. In this presentation, I will focus on two cases of using ETEM to give fundamental insights into surface reactions: oxidation and heterogeneous catalysis. Using a ultra-high vacuum ETEM, we have demonstrated that the transient oxidation stage of Cu and its alloys bear a striking resemblance to heteroepitaxy, where the initial stages of growth are dominated by oxygen surface diffusion. The second part of my talk will focus on heterogeneous catalysis, which depends sensitively on the nano-sized 3-dimensional structural habits of nanoparticles (NPs) and their physicochemical structural sensitivity to the environment. To exemplify synergistic combination of experimental tools (X-ray absorption spectroscopy, ETEM) coupled with theoretical simulations, we have shown that Pt NPs may be both ordered and disordered, depending on its size, support and adsorbates. A statistical description of nanoparticles is more appropriate in understanding structure/property of nanoparticles and their surface reactions. I will also highlight recent technical developments in this field, outline the current status as well as future needs and directions of high-resolution characterization methods, including combining spectroscopy and microscopy by using a versatile enclosed environmental cell that is compatible with both TEM and synchrotron sources.



## Beyond Crystallography: Atomic Resolution Electron Tomography

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Visualizing the arrangement of atoms has played an important role in the evolution of modern science and technology. Crystallography has long been used to reveal globally averaged 3D atomic structures. Scanning probe microscopes can determine surface structures at atomic level. Electron microscopes can routinely resolve atoms in 2D projections of 3D crystalline samples. In this talk, I will present a general method for 3D determination of *local* structures at atomic resolution. By combining scanning transmission electron microscopy with a novel data acquisition and 3D image reconstruction method, known as equal slope tomography (EST), we achieved electron tomography of a  $\sim 10$  nm Au nanoparticle at 2.4 Å resolution and identified several major grains in three dimensions (1). We also observed nearly all the atoms in a Pt nanoparticle and imaged, for the first time, the 3D core structure of edge and screw dislocations at atomic resolution (2). More recently, we determined the 3D coordinates of thousands of individual atoms and a point defect in a tungsten needle sample with a precision of  $\sim 19$  picometer, where the crystallinity of the sample was not assumed. From the coordinates of these individual atoms, we measured the atomic displacement field and the full strain tensor with a 3D resolution of 1 nm and a precision of  $10^{-3}$  (3). We expect this general atomic resolution electron tomography method to find broad applications in solid state physics, chemistry, materials sciences, nanoscience and biology.

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## Polarization-dependent Imaging of Domains, Critical Phenomena, and Dynamics using X-Ray Photoemission Electron Microscopy

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X-ray Photoemission Electron Microscopy (X-PEEM) is a surface imaging technique, which uses photoelectrons and secondary electrons created by the absorption of x-rays. A resolution of a few ten nanometers is typical for X-PEEM while newest, aberration corrected instruments promise to reach low nanometer resolution. Element-, chemical-, local symmetry-, and magnetic-contrast imaging exploit the tunability, high energy resolution, polarization control, and stability provided by modern x-ray beamlines. While not brightness limited, focusing of the x-rays allows us to reach high frame rates in X-PEEM experiments.

In the first half of this presentation I will discuss current trends in the technical development of X-PEEM, in particular aberration correction, cryogenic imaging, imaging of fast dynamics, and time-lapse imaging using fast CCD detectors. Today, the X-PEEM technique is being applied to a wide variety of materials and scientific problems and flexibility in regard to the environment, to temperature, magnetic fields, electric signals, and spectroscopic probes is as important as the spatial resolution of the instrument.

In the second part I will showcase several science examples that exploit polarization-dependent contrast. Ferroelectrics and multiferroics are seen as tools to control magnetism using electric fields, drastically reducing the energy required to switch a magnetic element and providing a direct, localized handle to the magnetic state. Starting with first experiments that demonstrated the ability of X-PEEM to measure the magnetic and ferroelectric domain state, I will show how PEEM imaging allowed researchers to build and characterize more and more sophisticated multiferroic systems, which bring us closer today to controlling nanoscale magnetic moments in a capacitive device that requires no currents [1,2].

Linear dichroism imaging has had great influence on our knowledge of the antiferromagnetic domain state but it is equally useful in structural studies with sub-micron spatial resolution. Using polarization dependent X-PEEM, researchers were able to disentangle the chemistry and orientation of crystallites in calcium carbonate biominerals, giving us important information about the morphology and growth of these materials. I will present some of the latest results on natural materials existing in shells and tunicates [3].

The ability of X-PEEM to rapidly map large areas with high resolution has been used in multiple studies of fluctuations and relaxations in artificial spin ice systems. Artificial spin ice systems that are made of nanoscale magnetic patterns are thermodynamical model systems that replicate frustrated lattices of atomic magnetic moments, for example of pyrochlore spin ice. X-PEEM imaging has been instrumental in allowing us to go beyond static imaging of glassy spin ice towards measuring thermally active systems that relax and fluctuate on time scales of milliseconds to hours [4].

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## Hard X-ray Microscope at 10 nm

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The Hard X-ray Nanoprobe (HXN) at the NSLS-II, constructed to achieve the world's most ambitious goal for hard x-ray microscopy, is now ready for general user experiments. The designed microscopy capabilities include measurements of specimen's morphology, composition, crystalline ordering, and chemical states. Intensive R&D for nanofocusing optics [1-3] and nanopositioning [4], together with innovative methods for achieving excellent beam stability, resulted in initial scanning microscopy capability with ~15 nm resolution. The presentation will elaborate on important instrument design features and commissioning experiment results.

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## X-ray Nano-imaging at ID16-NI, ESRF

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### Introduction and Objectives

Driven by the need to address more scientific challenges in energy, health, advanced materials and the environment, the European synchrotron ESRF has developed within its Upgrade Program a brand new nano-imaging end station ID16A-NI. This beamline is optimized for ultimate hard X-ray focusing with a large energy bandwidth at specific energies of 17.05 and 33.5 keV.

This new facility aims at the quantitative 3D characterization of the morphology and the elemental composition of specimens through the combination of coherent imaging techniques [1,2] and X-ray fluorescence analysis [3]. The applications in biomedicine, life sciences and nanotechnology will span over a large range of length scales which goes from the organelle level, over small organisms, nano- and micro-electronics to biopsies of mineralized tissue.

### Results and Discussion

Hard X-ray phase nanoscale holographic-tomography with a projection microscopy geometry has been implemented as a multi-scale approach for 3D imaging. A focal spot down to around 20nm with a flux up to  $7 \times 10^{11}$  photons/second has been achieved. With the a fast zoom-in and zoom-out from a few hundred nm down to a few nm, nano-tomography offers quantitative 3D structural information acquired from the retrieval of the electron density distribution of the objects.

Combined with X-ray fluorescence microscopy (XFM) in 2D/3D, correlative X-ray microscopy can quantify the complementary information of both structures and elements within whole cells with a probe of a few tens of nm in both imaging methods.

As another coherent imaging method, far-field ptychography has also been in development at the beamline to reach the highest spatial resolution the beamline can offer, also with the possibility to acquire simultaneously the fluorescence signals.

### Conclusions

The new hard X-ray imaging end station ID16NI at ESRF has been developed to achieve nano-focusing at two discrete high energies: 17.05 and 33.5 keV. Specialized in the coherent imaging techniques and XFM, also with cryo-environment to be added in the future, this beamline will pave the way to explore new potentials in biomedicine, bioengineering, and nanotechnology.

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## The use of X-ray micro- and nanoprobe to understand the natural degradation of semi-conductor materials used as artists' pigments

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Semiconductor materials such as HgS (vermillion red), CdS (cadmium yellow), As<sub>2</sub>S<sub>3</sub>/As<sub>4</sub>S<sub>4</sub> (orpiment/realgar) and Pb<sub>3</sub>O<sub>4</sub> (minium or red lead) as well as colored materials showing similar properties (such as PbCr<sub>1-x</sub>S<sub>x</sub>O<sub>4</sub>, chrome yellow) have been frequently used as artists' pigments in several historical periods. Depending on the physicochemical conditions of conservation, these materials can be subject to natural degradation reactions which may lead to noticeable changes in color and/or mechanical integrity of the paint layers or paintings in which the pigments were used. In general, both physical and chemical agents can be identified as the cause of such unwanted transformations; usually (but not always) the simultaneous presence of several risk factors is required to allow these reactions to proceed at a noticeable pace. Light of different wavelengths and cyclic variations in relative humidity are among the most important physical agents contributing to paint degradation. In the category 'chemical agents', next to the components of the paint itself (binding medium, filler materials, other admixture pigments), also external chemicals can be identified as being the cause of alteration reactions. Sometimes these are very innocuous in nature such as NaCl (or related) particulates deposited on the painting surface or atmospheric CO<sub>2</sub>. In several specific cases, we have observed that the degradation reactions not only involve a single-step chemical transformation, but may be comprised of (a) an initial photo-induced redox reaction followed by (b) some form of transportation or diffusion process of the initial secondary reaction products from the depth of the artifact towards the surface, which may optionally be followed by (c) other redox and/or precipitation reactions. Thus, pigment alteration frequently gives rise to strongly heterogeneous, layered materials of which the interlinked phases must be separately characterized on the micro- and nanoscale to allow for a better understanding of the sequence of reactions constituting a complete alteration pathway.

In this contribution, an overview of such recently investigated, spontaneously occurring transformations will be given, with emphasis on how synchrotron X-ray micro- and nanoprobe (and related) methods can be exploited to extract information from a limited number of paint micro samples. As specific cases, the degradation of HgS, CdS, Pb<sub>3</sub>O<sub>4</sub> and PbCr<sub>1-x</sub>S<sub>x</sub>O<sub>4</sub> in works by Rubens and Van Gogh will be discussed [1,2,3]. Next to micro- and nano-analytical methods we have also found it very useful to employ electrochemical methods to monitor the redox transformations of simplified semi-conductor pigment/paint samples under various well controlled physico-chemical conditions [4]. Such information also facilitates the use of theoretical prediction models for the reactivity of (pairs of) photo-sensitive semi-conductor materials.

A final aspect to which attention will be given is the fact that the insights related to chemical alterations, usually obtained from a small number of paint micro samples of a given artefact, of which the representativeness may vary, can be put in the larger context of the entire work of art by combining results from micro- and nanoscopic (X-ray based) analyses with macroscopic (X-ray based) imaging information.

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Oral Presentation Abstracts

# New Development of Silicon Drift Detectors and Readout Electronics for High-resolution and High-count Rate X-ray Spectroscopy

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## Abstract

The purpose of this presentation is to review the current status and possible future trends in the development of Silicon Drift Detectors (SDDs) and related electronics for high-resolution and high-rate X-ray spectroscopy. For several applications, in particular in microanalysis and in experiments at synchrotron light sources, SDDs have become the detector of choice, also with different commercial options available. However, the performances of the available SDD-based detection systems is further challenged by the need to further increase the counting rate capabilities, still keeping good energy resolution. This is particularly true for the ongoing upgrades of actual synchrotron light machines or for the use in future sources where a factor between 10 and 100 to beam-on-sample fluxes may be increased with respect to present conditions. More commonly used SDDs-based systems are usually limited to single- or few-channel systems (4-6 units). This motivates further developments of this technology toward multi-channels systems (e.g. 16-24 or even larger number of units) possibly based on a modular approach to build versatile systems for synchrotron applications, with also the advantage of an easier and cheaper replacement of malfunctioning units. New developments based on this approach will be presented in the talk. Regarding the front-end electronics, which covers a fundamental role in the noise performances, therefore on achievable energy resolution, the recent availability of CMOS preamplifiers to be directly connected to the detector as replacement of conventional JFETs allows an improvement of performances, in particular in the direction of high-count rate spectroscopy (e.g. 1Mcounts/s/channel). As an example, and energy resolution FWHM at 5.9keV of 126eV and 140eV has been measured with CUBE preamplifier at an analog shaping time of 250ns and digital peaking time of 100ns, respectively. The capability to obtain a satisfactory resolution at very short processing time provides the additional benefit to potentially relax the cooling conditions for the detector, as the contribution to the resolution due to the leakage current is less and less significant at short processing times. This opens the possibility to operate SDDs with a resolution better than 150eV even close to room temperature, with potential simplification of the detector apparatus.

For what concerns processing electronics, the push towards high-rate spectroscopy measurements further justifies the use of digital pulse processors in the detection system and commercial solutions are available (e.g. XSPRESS and Falcon systems). However, the perspective to build detection systems with few tens, maybe few hundreds, of readout channels, make the use of analog readout ASICs still of interest.



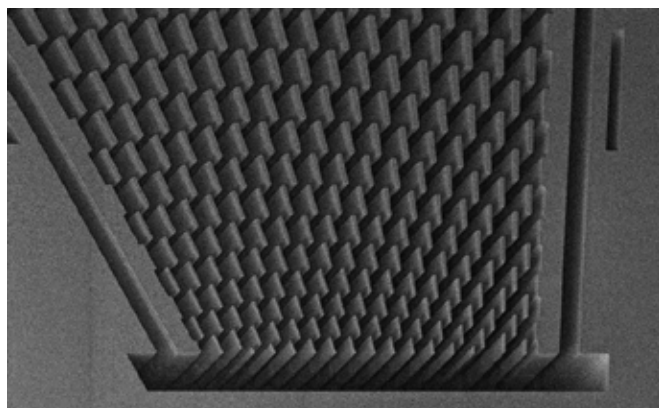
## Spoked Channel Arrays for 3D Micro Confocal X-ray Fluorescence

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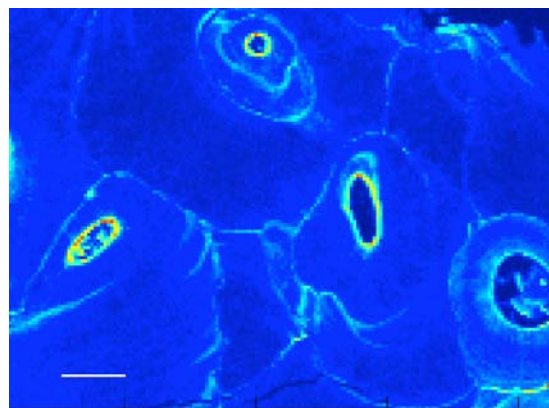
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Confocal X-ray Fluorescence Microscopy (CXRF) employs overlapping focal regions of two x-ray optics—a condenser and collector—to directly probe a 3D volume in space. In general, polycapillaries are used as the collector owing to their large solid angle of collection. Here, we report the practical demonstration of spoked channel arrays (SCAs), a novel x-ray collection optic for confocal x-ray fluorescence microscopy (CXRF). The optic consists of micron-scale, lithographically-fabricated arrays of collimating channels (Fig. 1), all directed towards a single source position. In contrast to polycapillaries, the spatial resolution of these optics is nearly energy-independent. Most recently, we have fabricated a set of optics made from germanium substrates, and successfully demonstrated (Fig. 2) their operation at 3- $\mu\text{m}$  and 7- $\mu\text{m}$  depth resolution from 2-20 keV. Among other benefits, this allows direct, 3D mapping of light elements such as P, K and Ca at the micron scale for the first time. The feasibility of these optics for confocal XRF and XAFS on a variety of different sample types were recently tested at APS beamline 20-ID-B, where they are now available to general users. The optic mounts in a custom-built holder, designed to mate easily to a single-element Vortex EX detector. Opportunities for future development will be discussed, including operation up to 30 keV, and operation in conjunction with pixel-array-based energy dispersive detectors, such as the Maia. The later combination would allow fast, 3D mapping of samples not suitable for computed tomography, and suggests the possibility of simultaneous, parallel detection of emission from different depths in a sample, e.g. for fast 3D mapping of major elemental constituents, or time- and depth-resolved, in-operando studies of chemical reactions in layered systems.



**Figure 1:** Scanning electron micrograph of a lithographically-patterned channel array.



**Figure 2:** 2D map of Pb  $L\alpha$  fluorescence in an un-thinned bone sample, obtained using confocal XRF with a spoked channel array as the collection optic. Scale bar is 100  $\mu\text{m}$ .

# High Speed X-ray Imaging and Spectroscopy with novel direct detection CMOS based Active Pixel Sensors

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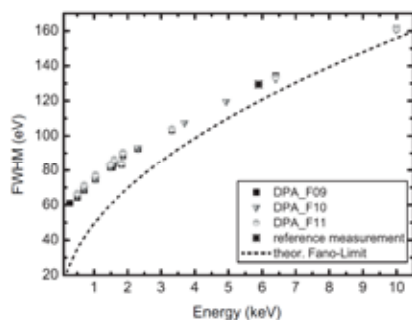
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## Introduction and Objectives

High speed counting, imaging and spectroscopic measurements of X-rays at energies between 100 eV and 30 keV require detectors with challenging properties. A back-illuminated 450  $\mu\text{m}$  thick active pixel sensor (APS) was developed and qualified for planetary science applications. Matched to the resolution of the X-ray optics aboard it has 300  $\mu\text{m}$  x 300  $\mu\text{m}$  large pixels in a format of 64 x 64, resulting in a sensitive area of 2 x 2  $\text{cm}^2$ . It is operated at a frame rate of 7.000 Hz, still delivering Fano limited imaging X-ray spectroscopy. Because the readout is so fast (30 Megapixel per second), the cooling requirements are relaxed and operation can be performed with a thermoelectric cooler. The new device is based on the DePFET principle built on fully depleted back-illuminated 450  $\mu\text{m}$  high resistivity silicon.

## Results and Discussion

To speed up the readout, the sensor is subdivided in two hemispheres, which are read out in parallel. As the regions to be read out can be programmed, areas with higher X-ray flux can be read out more frequently than areas with lower intensity. We have tested the detector system operated at full speed at the calibrated X-ray end stations SX700 and KMC of the PTB (Physikalisch-Technische Bundesanstalt, the German institute of standards) at the synchrotron BESSY in Berlin. Fig. 1 shows a summary of three different detectors systems. The X-ray energies were ranging from 250 eV up to 10 keV. The intrinsic physical limit in energy resolution given by the ionization statistics described by the Fano factor is shown as well. At 200 eV the energy resolution is 61 eV (FWHM) and it is 161 eV (FWHM) at 10 keV. Three different detector systems were tested, they are behaving quite similarly. This concept allows shrinking the pixel size down to 20  $\mu\text{m}$ . Beside X-ray imaging and spectroscopy the detector is equally suited for electron detection from 1 keV up to 400 keV. As in this case a higher readout noise can be tolerated the frame rate can be increased to more than 20.000 frames per second.



**Fig. 1:** Measured energy resolution from 200 eV up to 10 keV for three sensor systems. The dashed line is the theoretical resolution limit given by the ionization statistics. The symbols above are the measured data from the DePFET camera. The X-ray spectra were taken in a single photon counting mode at the PTB end stations SX700 and KMC at the BESSY synchrotron in Berlin with 7 000 frames per second.  $30 \times 10^6$  pixels are read out in the high energy resolution mode.

## Conclusions

A new high resolution high speed imaging spectrometer is shown, operating close to the theoretical Fano limit, having a monolithic size of 4  $\text{cm}^2$ . The quantum efficiency is above 90 % from 500 eV up to 12 keV. About 30 million pixel are read out per second with a read noise below 3.5 electrons (rms). More operational options and further improvements of the new concept will be shown.

## Multielement Amplifier Readout System for Silicon Drift Detectors

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Silicon diode and Silicon drift detectors (SDDs) have widespread applications in astrophysics, high energy physics, crystallography and medical imaging. Maia, a scanning x-ray fluorescence microprobe was designed with a multi-element photodiode sensor with 384 pixels and two application specific integrated circuits (ASICs) [1,2,3]. The detector was wire-bonded to a front-end ASIC which provided low noise charge pre-amplification and shaping. The output of the shaper was wire-bonded to a precision peak and time detector ASIC with arbitration logic and analog memory.

We present a new Multi-element Amplifier Readout System (MARS) ASIC that has been designed and fabricated in 250 nm CMOS to replace the ASICs currently used in the Maia x-ray microprobe system. The (MARS) ASIC instruments electrons or holes and combines the functions of the two chips currently used for Maia while significantly reducing the physical footprint, the electronic noise, and the power dissipation. The ASIC dissipates 127 mW and combines 32 channels. Each channel is comprised of low noise charge amplification, high order shaping with baseline stabilization, discrimination, pileup rejection, and peak and time detection with analog memory. The readout is sparse with the channel address. Any channel with a processed above threshold event can be read out independent of the other processing channels. The interface is analog differential and digital LVDS. Preliminary measurements for a 96-element silicon detector array will be presented.

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## X-ray Imaging of Tumor Microvasculature

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### Abstract

We show that optimized Au nanoparticles enable high resolution three dimensional (3D) x-ray imaging to detect the angiogenic microvessels of brain glioma tumors and the related leakage. The study included in vivo and post-mortem whole organ phase contrast imaging plus nanoscale 3D X-ray microscopy. This strategy led to a detailed analysis of the relation between xenografted cancer cells and the tumor-induced angiogenic microvasculature in mouse brains. In addition, we could detect and study nanoparticle leakage, a phenomenon of interest for nanomedicine applications. Specifically, we were able to analyze tumors induced by stereotactic inoculated glioma cells, and to correlate their position and propagation to the microvasculature. Complementary tests were conducted on cells loaded with fluorescent Au nanoparticles. We thus found that, after endocytotic internalization, such nanoparticles allow visible-light detection of cells. Furthermore, they exhibit the same contrast-agent performances as non-fluorescent nanoparticles, becoming dual imaging agents.

## Three-Dimensional Microstructural Imaging and Analysis of Energy Materials

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### Abstract

Materials used in energy storage and conversion (e.g. fuel cells, batteries, electrolyzers, CO<sub>2</sub> separation membranes) all consist of heterogeneous functional materials that exhibit functional behavior in a manner that controls their collective performance as an energy system. There is a critical need to understand the role of a material's structure, morphology, and composition on system performance<sup>1</sup>. This talk presents a non-destructive approach to image and characterize energy materials using synchrotron-based transmission x-ray microscopy, including XANES nanotomography<sup>2</sup> to analyze chemical and structural changes in materials during operation, and high temperature in situ imaging of Ni particles during oxidation and reduction<sup>3</sup>. Several theoretical approaches including numerical methods (lattice Boltzmann, finite element<sup>4</sup>) and analytical methods (electrochemical fin theory<sup>5</sup>) are used to analyze mass transfer, heat transfer, ionic/electronic charge transfer, and chemical / electrochemical reaction rates. To demonstrate this approach, solid oxide fuel cell and ceramic composite gas separation membranes are examined to provide fundamental insight into the origins of microstructured-induced transport losses during operation.

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### About the Speaker

Wilson K. S. Chiu earned his M.S. and Ph.D. degrees in Mechanical Engineering from Rutgers University in 1997 and 1999, respectively. His research was supported by the U.S. Department of Energy, National Science Foundation, Office of Naval Research, Army Research Office, and industry. He published 96 journal articles and 148 conference articles/abstracts. Among his honors, he was elected fellow of ASME in 2013, into the *Connecticut Academy of Science and Engineering* in 2013, received the *Rutgers University School of Engineering Medal of Excellence Award for Distinguished Young Alumnus* in 2010, the *United Technologies Corporation Professorship in Engineering Innovation* in 2008, and the *ASME Bergles-Rohsenow Young Investigator Award in Heat Transfer* in 2006. Starting on July 1, 2015, he will serve as the Editor of the *Journal of Fuel Cell Science and Technology*. He also serves on the editorial board of *Scientific Reports*, as an associate editor for the *ASME Journal of Heat Transfer* and the *International Journal of Thermal Sciences*, executive committee co-chair for the *ASME Advanced Energy Systems Division*, and executive committee member for the *ECS High Temperature Materials Division*. He has given over 90 plenary, keynote and invited lectures in the United States and abroad.

## Morphological and Chemical Evolution of Nanoporous Metal using X-ray Nano-tomography and Hard X-ray Spectroscopic Imaging

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Advancing technologies in energy storage and conversion devices such as batteries and fuel cells depends on developing novel materials. In particular, nanoporous materials—also known as nanofoams—exhibit unique properties such as high surface-to-volume ratio and a continuous network, which enables transport of fuels or electrolyte. A new dealloying method based on utilizing metallic melt as the dealloying agent instead of an aqueous solution has led to the successful fabrication nanoporous metal from less-noble metals, for example, stainless steels. This greatly reduces the cost of the material, while preserving the unique morphological factors that are desirable for energy applications. It is, however, critical to establish the correlation between the processes, morphology (2D & 3D) and chemical heterogeneity of these novel nanoporous metallic materials for suitable applications.

We utilized full-field nano-tomography via transmission x-ray microscopy to study the evolution of the morphology as a function of processing conditions, including precursor compositions, dealloying temperature and time. A systematic trend of spontaneous coarsening as a result of prolonged dealloying time and increased temperature was observed: the precursor compositions significantly impact not just the porosity of the nanoporous materials, but also the surface shape of the nanofoams. A gradient of the ligament size and compositional change along the dealloying direction were also observed. In addition, spectroscopic imaging at synchrotron sources is a powerful technique for spatially resolving chemical and elemental distributions in these materials. Utilizing the Sub-micron Resolution X-ray Spectroscopy (SRX) beamline at NSLS-II provides additional elemental and chemical information of the nano-porous materials.

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## Revisit of wetting on soft solids by TXM

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Many natural and synthetic materials are soft and deformable owing to the nature as in rubbers or to their structure as for thin films and fibers. Wetting behaviour of soft deformable solids, that is, elastowetting, is a very old subject and much attention has been paid over the past half century for various scientific issues involved in soft tissues and polymer gels, as well as for practical applications such as inkjet printing and microfluidic devices. One of the most questionable issues in wetting is the force balance that includes the vertical component of liquid surface tension. On soft solids, the vertical component leads to a microscopic protrusion of the contact line, that is, a ‘wetting ridge’. The wetting principle determining the tip geometry of the ridge is at the heart of the issues over the past half century. Here we reveal a universal wetting principle from the ridge tips directly visualized with high spatio-temporal resolution of Transmission X-ray microscopy (TXM). We find that the cusp of the ridge is bent with an asymmetric tip, whose geometry is invariant during ridge growth or by surface softness. This singular asymmetry is deduced by linking the macroscopic and microscopic contact angles to Young and Neuman laws, respectively. We also discuss dynamic wetting behaviors on soft viscoelastic solids that are important to interpret complex biological processes resulted from cell-substrate interactions. Our finding shows that this dual-scale approach would be contributable to a general framework in elastowetting, and give hints to issues in cell-substrate interaction and elasto-capillary problems.

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## 2D slope surface profilers based on a Shack Hartmann Sensor for sub 100 nrad slope error measurement

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Focusing high brightness x-ray light available with third/fourth generation synchrotron/FEL sources requires significant advances in the quality of X-ray mirrors and in the metrology used to optimize their fabrication. The operational solution from NSLSII and Imagine Optic (a French optics technologies company) was to develop a way to perform automated high accuracy optical measurements for the testing of advanced x-ray mirrors. The adopted solution is a piece of equipment based mainly on a Stitching Shack Hartmann Optical Head (SSH-OH). This approach is innovative and provides a stand-alone, non-contact, large-surface high accuracy optical metrology system for 2D slope measurements. We will describe the measurement setup and discuss the results obtained with the developed system.

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## Kinoform Lenses for High Energy (>50 keV) Photons and other applications

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Many applications, especially the study of high Z materials, benefit from the use of high-energy x-ray photons (>50 keV) from high brightness accelerator based sources. Focusing optics for such high energies are not widely available, and do not have the quality that is commonplace at lower photon energies (<10 keV). Using silicon kinoform lenses at beamline 1-ID at APS with 51 keV photons, we obtain 1 dimensional focal spot sizes of  $225 \pm 25$  nm at a focal length of 0.25 m with a gain of  $87 \pm 4$ , and a spot size of  $1 \pm 0.1$  microns at a focal length of 2 m with a gain of  $176 \pm 5$ . A point focus was also obtained with a crossed kinoform geometry, similar to the KB mirror geometry, and a point focus of 0.85 microns (v) by 2.3 microns (h) was measured. At a higher energy of 102 keV, we obtained a 1.5 micron spot size. We anticipate that these optics will enable many x-ray microscopy techniques at high photon energies. Finally, we also discuss some x-ray micro-diffraction results obtained on human hair using kinoform lenses at 12.1 keV.

## High efficiency polymer lenses for hard X-ray nanofocusing

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Nanofocusing, adiabatic, polymer compound refractive lenses combine the best of two worlds: the ease of alignment – common for refractive X-ray lenses – and a large effective aperture combined with a long working distance between exit aperture and focal plane even for hard X-rays – which is a feature usually attributed to reflective optics. This combination has long been desired in materials science related research. X-ray diffraction with nanofocused hard X-rays readily serves structural information with sub- $\mu\text{m}$  spatial resolution from crystalline and semi-crystalline materials (e.g. metals, biomaterials, synthetic compounds). That way grain orientation, residual stress profiles, crystal structure or texture can be obtained in a non-destructive analysis. Provided a long working distance focusing element, these high resolution nanodiffraction experiments can be performed in extended sample environments, with in-situ conditions and from strongly absorbing metallic samples.



Figure 1: (left) Crossed adiabatic CRL prototype, with a working distance of 43 mm at 13 keV, installed at P03 nanofocus endstation in 2015. (right) SEM image of a CRL with varying aperture, assembled from two half lenses under  $90^\circ$ .

The adiabatic lens system presented here has a large entrance aperture of  $170\ \mu\text{m}$  and a continuously decreasing aperture adapted to the local beam diameter. In this way the exit numerical aperture of the lens stack is maximized and the focal spot diameter is minimized. Refractive polymer lenses can be fabricated via deep X-ray lithography out of SU-8, a commonly used epoxy-based negative photoresist and can be operated in air at ambient conditions. A crossed geometry, intersected lens stack provides an efficient two-dimensionally focusing close to free of spherical and astigmatic aberration. These lenses have been proven to be long time radiation stable at multiple synchrotron radiation applications and in laboratory settings up to a deposited dose of  $2\ \text{MJ}/\text{cm}^2$  [1]. At PETRA III synchrotron radiation source, the lenses have been successfully used in full field microscopy setups (beamline P05, [2]) and for X-ray nanodiffraction experiments (nanofocus endstation of P03 beamline). At P03 the lenses were used to directly demagnify the undulator source at 13 keV, yielding a focal spot size of 500 nm at a clear working distance of 43 mm. These results met the design specification and required only a short initial alignment procedure. Due to an effective aperture of  $34\ \mu\text{m}$  the transmission of the utilized lens stack was 16 %.

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# Analysis of compartmentation and binding of metals in plants at physiological and toxic concentrations analysed in frozen-hydrated samples by $\mu$ XRF tomography and related techniques

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Various transition metals are essential nutrients, but toxic at elevated concentrations. Many mechanisms of metal uptake, transport, binding and sequestration are shared between animals (incl. humans), fungi, plants and even bacteria, others are specific for one group. Analyzing metal distribution (compartmentation) and speciation is a crucial step in revealing mechanisms of metal uptake, transport sequestration, deficiency, toxicity and detoxification. The least artifact-prone techniques for this task all belong to the field of X-ray spectroscopy, including EDX, PIXE, XRF and XAS. For minimizing artifacts, element re-distribution and ligand exchanges inside the measured tissues have to be prevented. The most reliable method for reaching this aim is measurement of shock-frozen, hydrated tissues. Ideally, these samples are analyzed as bulk tissues, replacing physical thin sectioning by recording  $\mu$ XRF tomograms [1-3]. In the ideal case the same samples can be analyzed by  $\mu$ XAS as well, allowing a direct correlation between element distribution and local differences in speciation [1]. Depending on the beamline, resolution of details even on the subcellular level is possible in such samples. In this talk, this ideal case will be compared to related techniques of measurement, in particular SEM-EDX [4-7] and XAS on powdered frozen-hydrated samples [8-12]. Alternative sample preparation techniques (e.g. conventional freezing in liquid nitrogen, freeze-drying and freeze-substitution) will be discussed as well. Differences between the techniques, especially in terms of artifact risks, will be highlighted.

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## Advances in Diffraction Tomography: deep sub-micrometer resolution

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### Introduction and Objectives

Performances and physical properties of high technology materials are influenced or even determined by their initial microstructure and by the behaviour of impurity phases. To characterize these impurities and understand their relations with the surrounding matrix, non-destructive techniques have to be used. Diffraction Tomography is a recent and powerful tool to analyze the 3D crystallographic composition of materials [1-3]. This technique was previously limited to samples giving a powder diffraction signal and had problems with coarse grained materials. Our developments overcame the granularity problem in order to reach sub-micron resolution when using an X-ray nanoprobe for diffraction tomography [4].

### Results and Discussion

The sample was a powder composed of U-Mo alloy particles (about 25  $\mu\text{m}$  in diameter) surrounded by a  $\text{UO}_2$  protective layer [5]. A particle core is made up of several U-Mo alloy grains with an average diameter smaller than 3  $\mu\text{m}$ . This core is covered by a 1  $\mu\text{m}$  thick  $\text{UO}_2$  protective layer (grains size  $\sim 10$  nm). The grain map for a 2D slice of the particle is shown Fig. 1. Due to the high resolution of this technique, a minority  $\text{U(C,O)}$  phase (1 wt. %) with sub-micrometer sized grains was characterized inside the particle. The onset of  $\text{U(C,O)}$  grain crystallization can be described by a precipitation mechanism since a single U-Mo grain has direct orientation relationship with more than one of the surrounding  $\text{U(C,O)}$  grains.

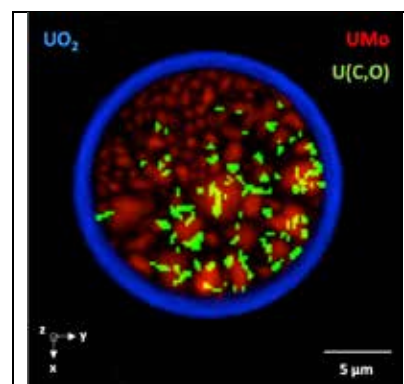


Fig.1: High resolution diffraction tomography slice of a U-Mo atomized particle.

### Conclusions

Due to advanced reconstruction techniques and an adapted acquisition scheme, XRD-CT can now provide both powder diffraction and single crystal type X-ray structure analyses combined with tomographic imaging.

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## Cryogenic Analysis of Frozen Hydrated Biological Tissue at the Hard X-ray Micro-Probe Beamline at PETRA III

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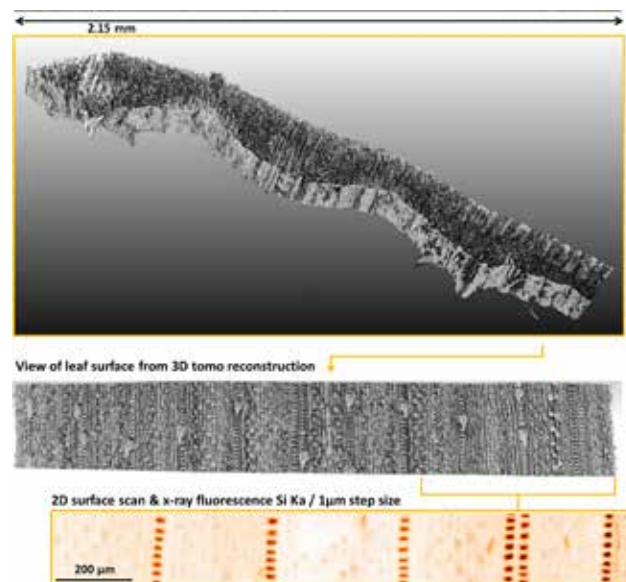
### Introduction and Objectives

A robust cryogenic workflow was developed at beamline P06 to minimise structural and chemical artefacts during sample preparation as well as beam damage during analysis. Shock frozen hydrated biological and medical tissue samples up to a diameter of 2 mm can be analysed. The developed instrument consists of a vacuum chamber (ca.  $10^{-8}$  mbar), two cooling systems, one for rapid cooling at startup and one vibration free long term cooling system maintaining the temperature between -120 °C and -150 °C, an ultrathin polymer window XRF detector, an absorption imaging detector (PCO 4000), and a cryogenic vacuum transfer system.

The presented system allows besides 2D and 3D elemental imaging also absorption and phase contrast tomography on the (sub-)micrometer scale.

### Results and Discussion

Recent results will be presented, demonstrating the capabilities regarding absorption/phase tomography and fluorescence imaging on frozen-hydrated biological samples. Analysis of a rice leaf shows the ability of the presented instrument to obtain high resolution 3D images, while the Si elemental image shows the high sensitivity and resolution of the XRF imaging system.



**Figure 1 (Top)** A crop of a 3D CT of a shock frozen hydrated rice leaf. **(Center)** Front absorption view **(Bottom)** A 2D XRF Si image, showing Si inclusions in the epidermis of a rice leaf.

### Conclusions

Combining a full cryogenic workflow with a cryogenic measurement instrument allows for high resolution structural and high sensitivity elemental analysis without the need of any chemical sample preparation steps, preserving the original state as much as possible.